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Novel Chemistry at the National Ignition Facility

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American Chemical Society, Denver

8/28/2011

Jon Eggert

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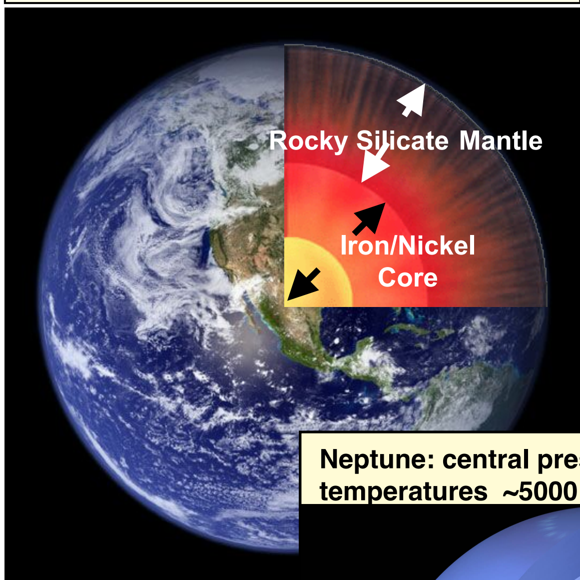
The National Ignition Facility (NIF) is currently a 192 beam, 1.3 MJ laser

This spring we performed the first materials experiments on the NIF, establishing NIF as a uniquely-capable platform for a new regime of extreme-compression science.

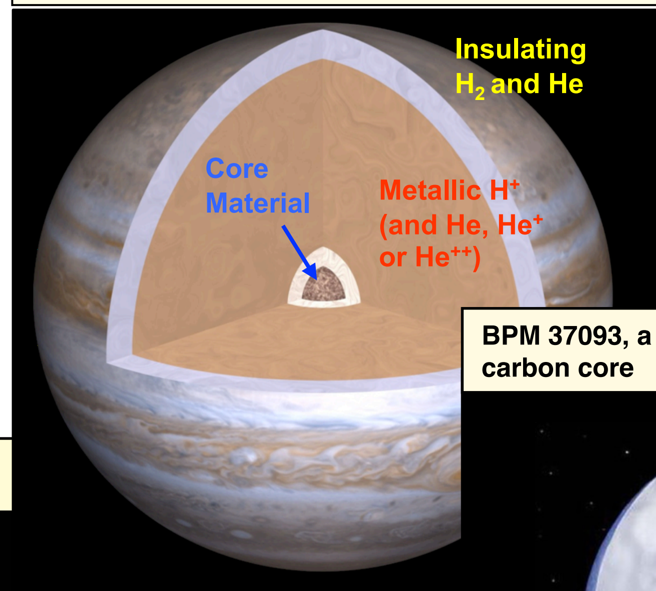
Target chamber with steel frameworks for catwalks being installed.

Matter at very extreme densities and temperatures is quite common in our universe

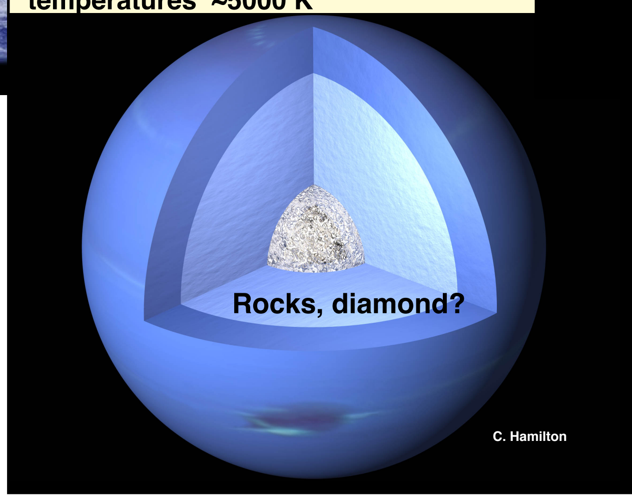
Earth: central pressures ~3.6 Mbar and temperatures ~6000 K



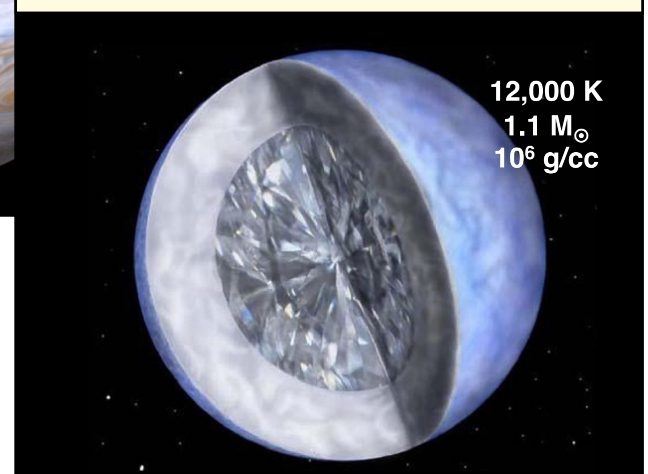
Jupiter: central pressures ~77 Mbar and temperatures ~16000 K



Neptune: central pressures 8 Mbar and temperatures ~5000 K



BPM 37093, a white dwarf star with a solid carbon core



"BPM 37093: A seismological test of crystallization theory in white dwarfs"
Kanaan, et al., A&A (2005)

We are considering pressures approaching the atomic unit of pressure (1 Hartree / cubic Bohr radius)

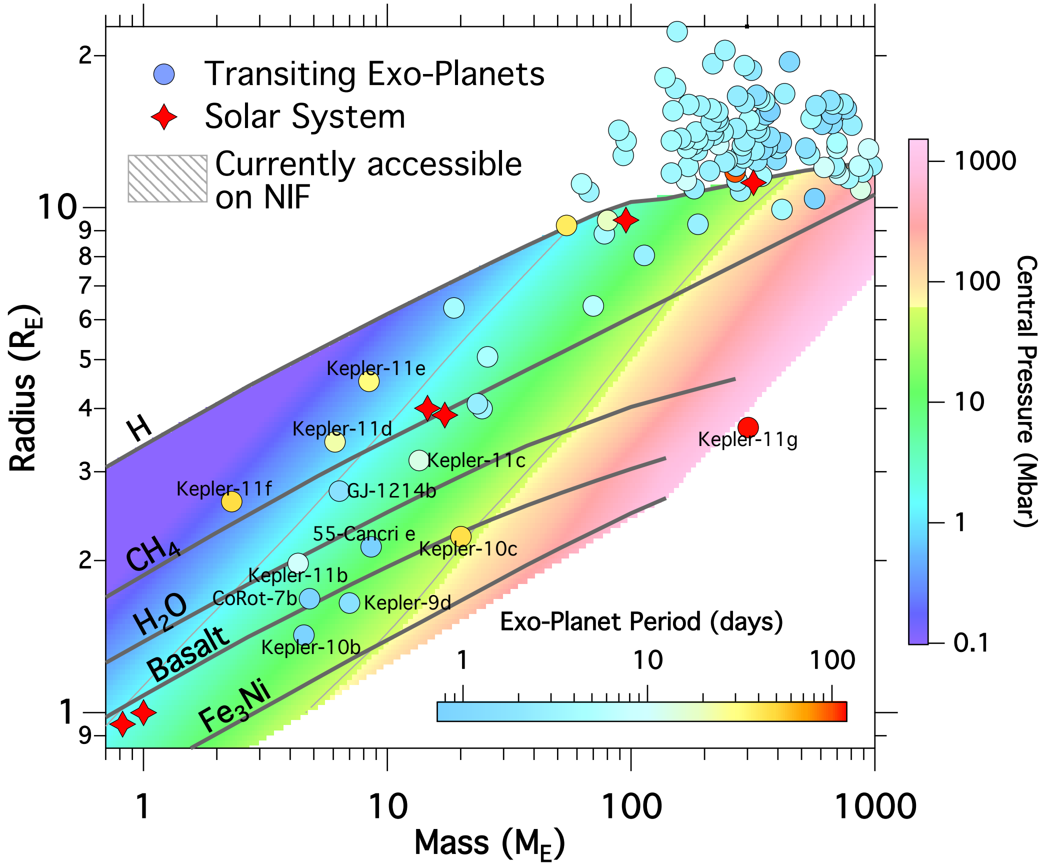
Date	Atomic unit		Discoverer	Capability/implication
1880				
	Energy	$E_h = m_e e^4 / \hbar^2 = 27.2 eV$	Rydberg	spectroscopy =>quantum mechanics
1900	Mass	$m_e = 9.11 \bullet 10^{-31} kg$	Thomson	mass spectrometry
	Charge	$e = 1.6 \bullet 10^{-19} C$	Millikan	oil drop =>atoms are divisible
1920	Length	$a_0 = \hbar^2 / m_e e^2 = .0529 nm$	Bragg	diffraction =>crystal structure
1940				
1960				
1980	Time	$t = \hbar / E_h = 27 \bullet 10^{-18}$	Krausz	attosecond spectroscopy =>observe electron bonding
2000				
	Pressure	$P = E_h / a_0^3 = 294 Mbar$	NIF	Fundamental change in matter from KeV chemistry to macro-quantized states
2020				

D. Hicks

The atomic unit of pressure is the pressure required to "seriously disrupt the shell structure of atoms" (Bukowinski,1994)

At least 573 exo-planets, including 144 transiting planets have been observed and confirmed*

*As of Aug. 11, 2011 -- http://exoplanet.eu/



Central pressures are interpolated, undifferentiated compositions and underestimate results for differentiated interior models.
Swift, et al. (2011)

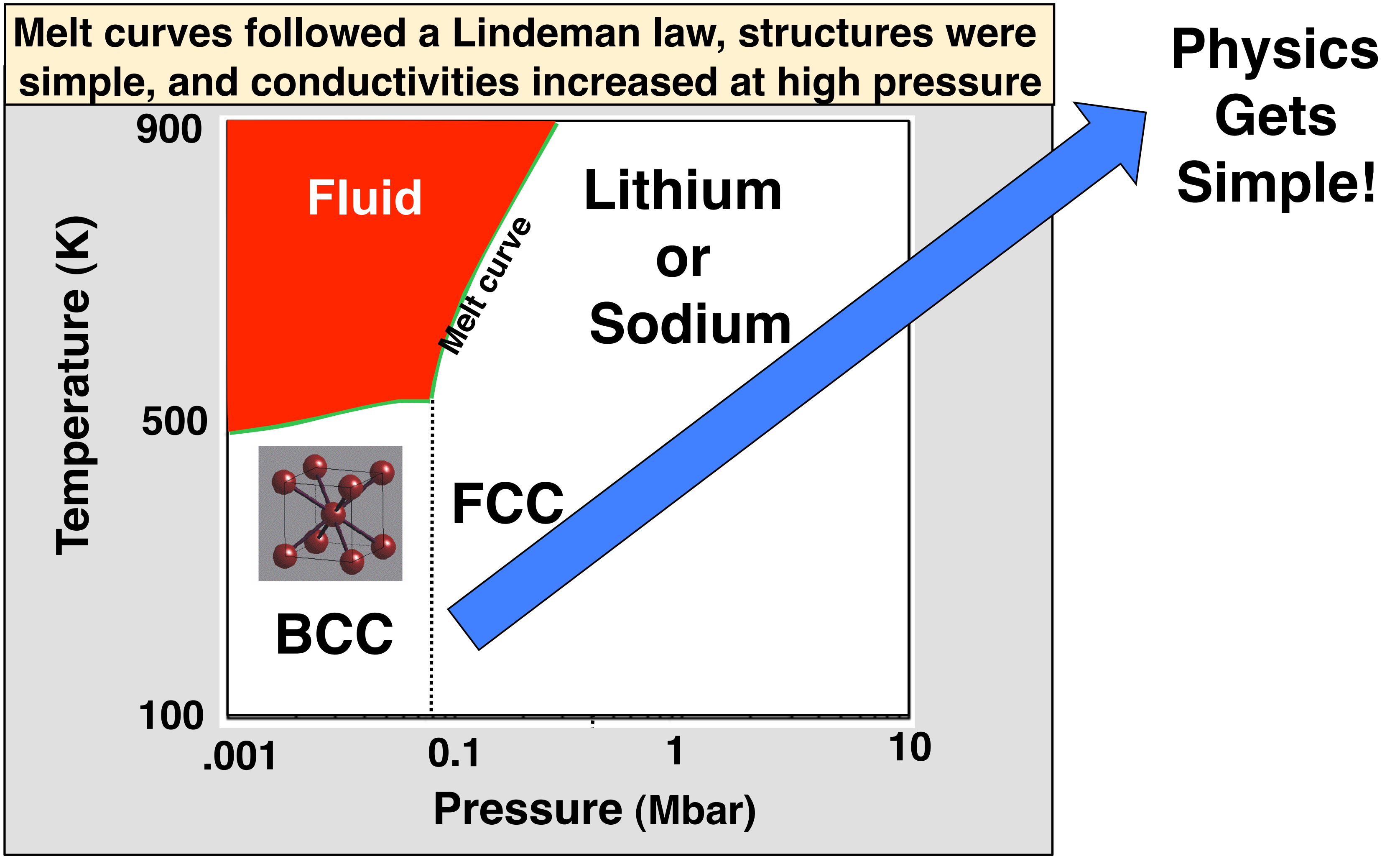
Heavy detection biases favor large Masses and Radii and small Orbits.

~7 rocky or metallic "super earths"
~2 "water worlds"
~6-9 "icy giants"

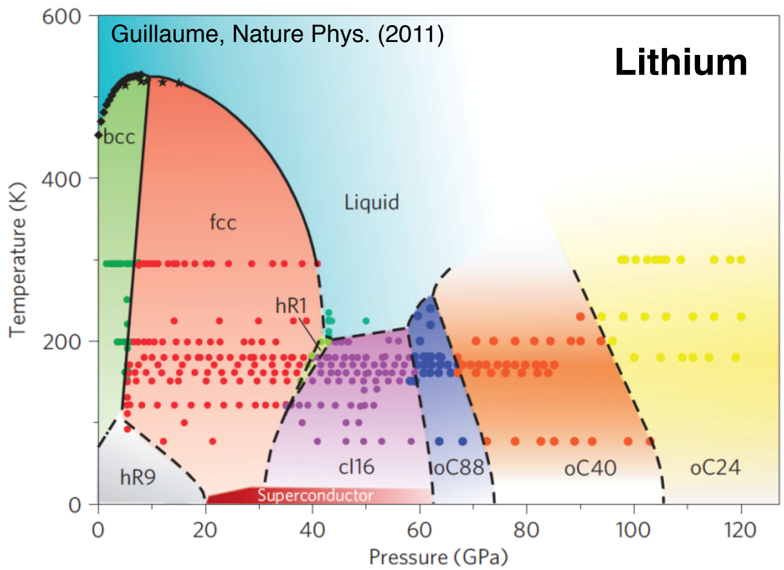
Central pressure estimates require an interior model and material EOSs

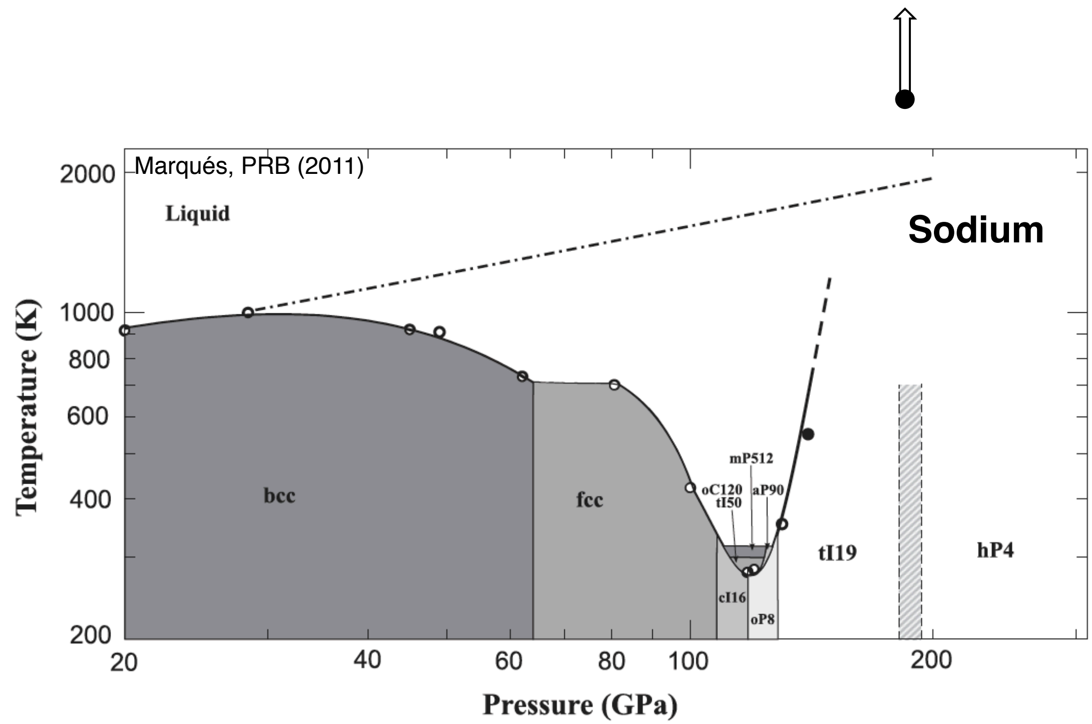
Material properties are needed at 10-1000 Mbar

Just a few years ago, ultra-high pressure phase diagrams for materials were very "simple"



Recent observations find complex high-pressure behavior





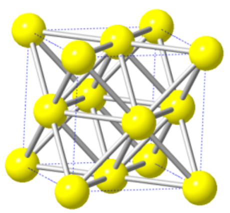
Melt curves and structures are experimentally very complex for alkali metals.

New experiments and theories point out surprising and decidedly complex behavior at the highest pressures considered.

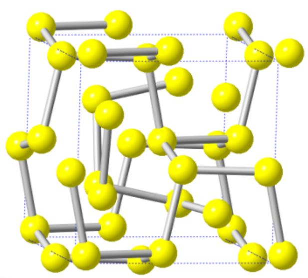
Traditional view: All materials become simple at high pressure appears to be incorrect!

> **“. . . what the present results most assuredly demonstrate is the importance of pressure in revealing the limitations of previously hallowed models of solids”**
> **–Neil Ashcroft (2009).**

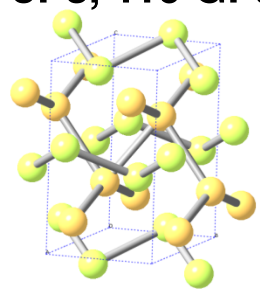
FCC, 65 GPa



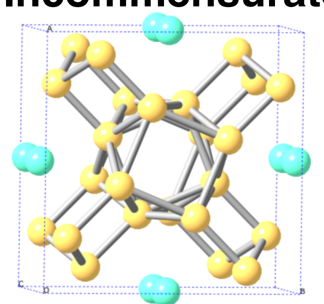
cI16, 108 GPa



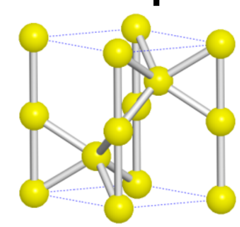
oP8, 119 GPa

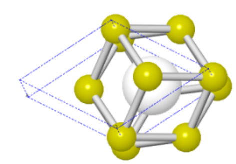


tI19, 147 GPa Incommensurate

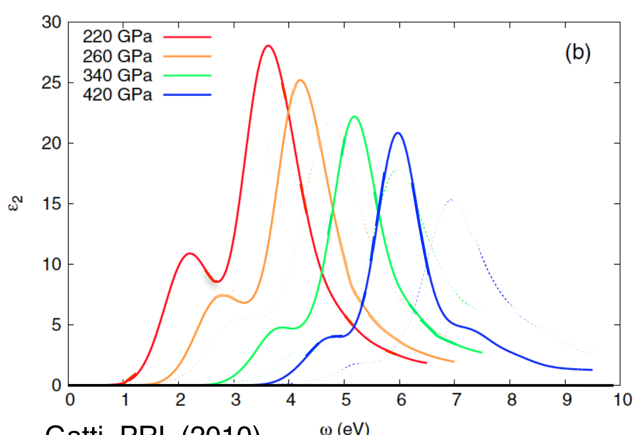


hP4, 190 GPa Insulating, Transparent Electride



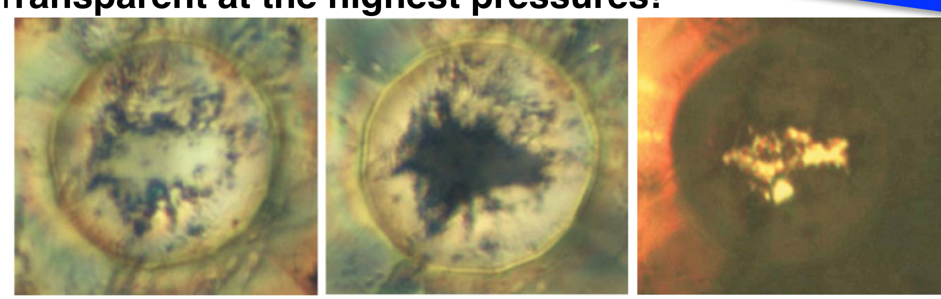


Increasing Structural Complexity



Gatti, PRL (2010)

Transparent at the highest pressures!



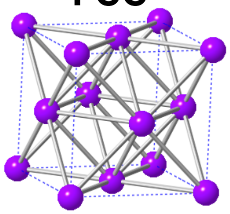
120 GPa 156 GPa 199 GPa

Ma, Nature (2009)

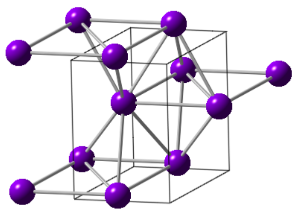
High pressures phases of aluminum are also predicted to be complex

Pickard and Needs, Nature Materials (2010).

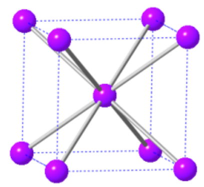
FCC



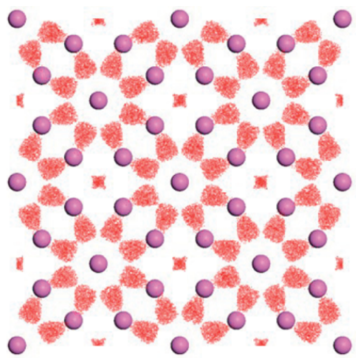
HCP
2.17 Mbar

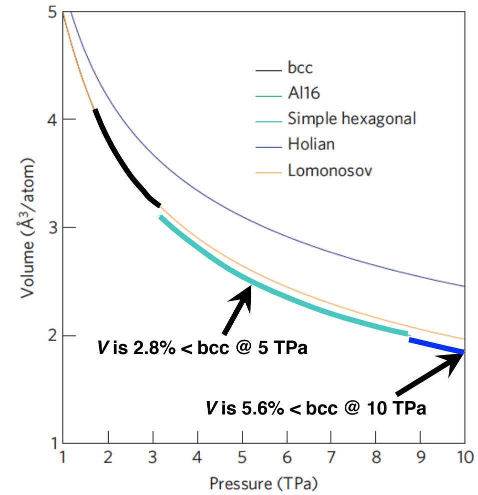


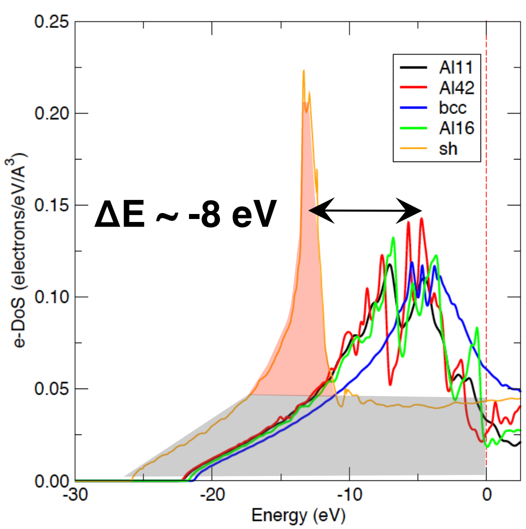
BCC
3.8 Mbar



Host-Guest structure of Ba-IVa (Incommensurate Electride)
32-88 Mbar



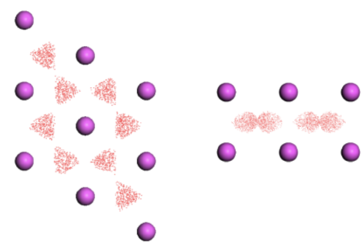




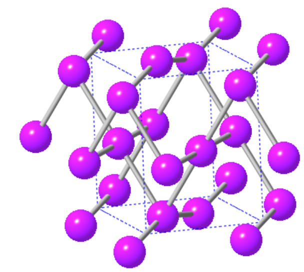
ΔV~2.8%

Simple Hexagonal Electride
88 – 100 Mbar

ΔV~1.8%



CMMA Electride
> 100 Mbar

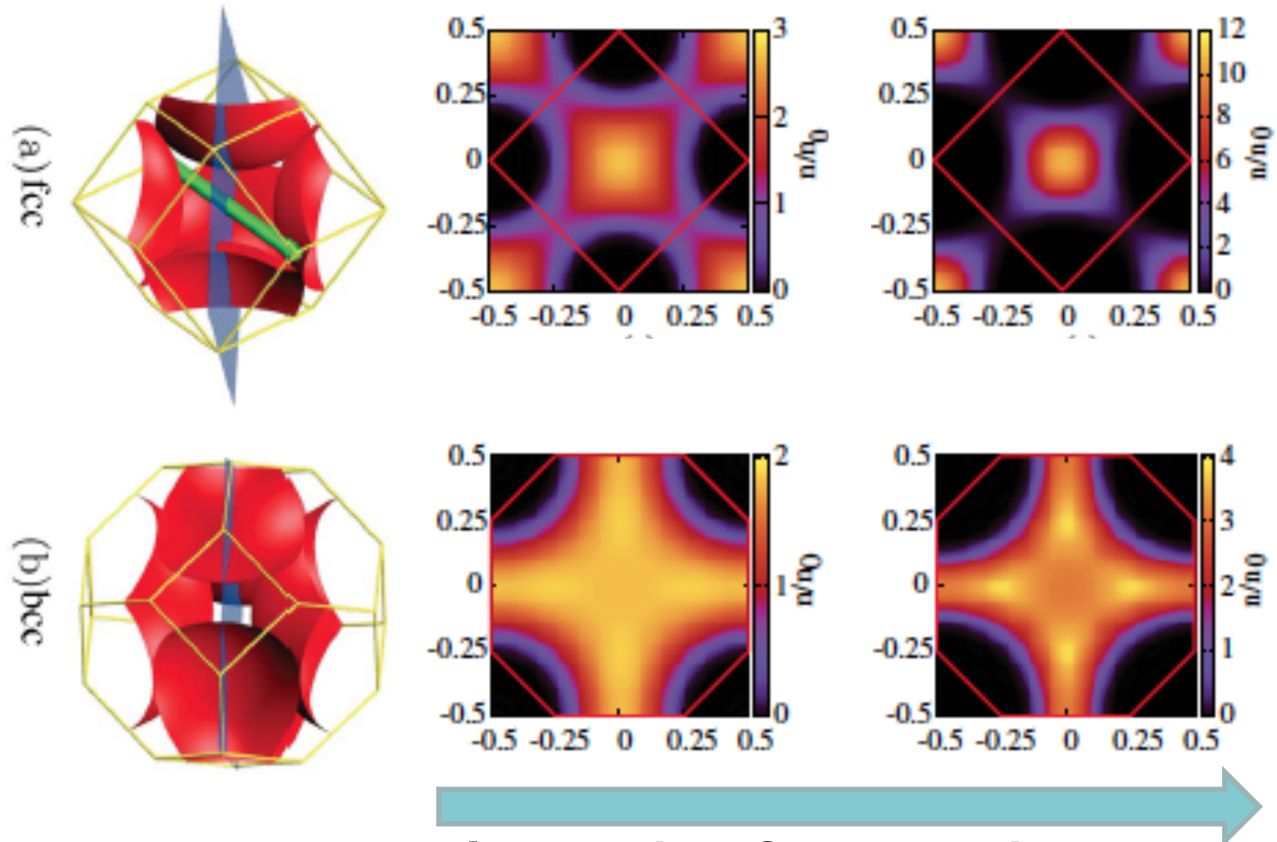


No predictions yet for melt line of aluminum

“all structures near 300 Mbar are far from close packed”

Fundamental Quantum-Mechanical driving forces appear to be responsible for this complexity at extreme compression

Hard-sphere potential with Pauli exclusion and orthogonal wave functions



Increasing Compression

"When the cores are induced to occupy an increasingly larger fraction of the unit cell the indications are that *a new paradigm*, as suggested here, may be appropriate."
--Rousseau and Ashcroft, PRL (2008)

A new paradigm. Really?

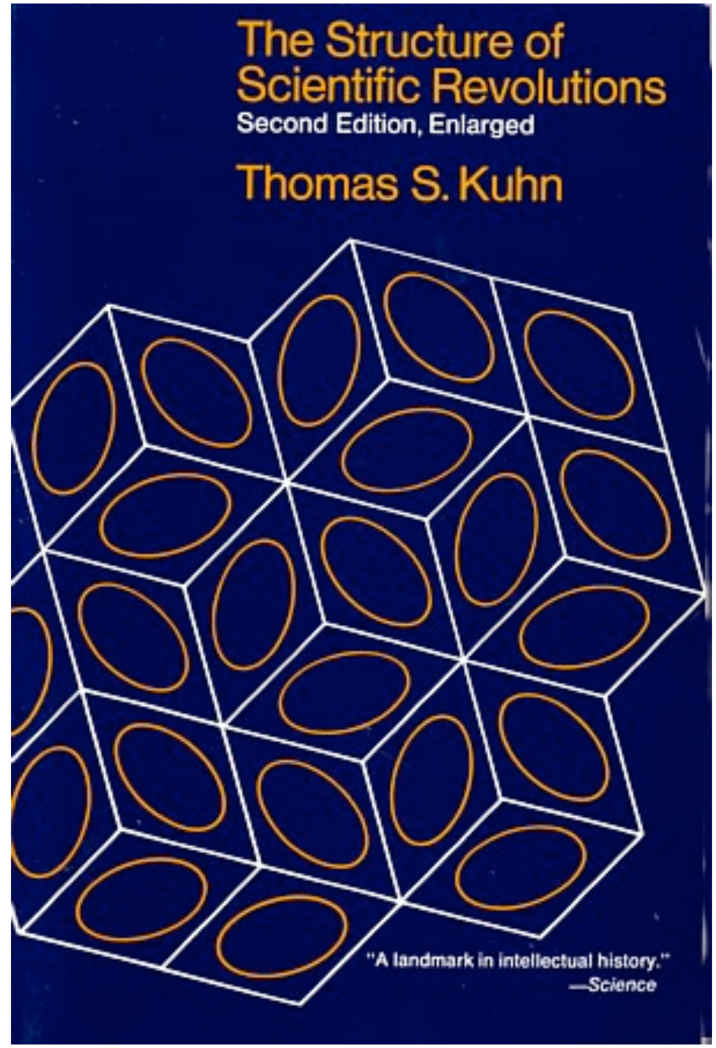
Are we really about to witness a true paradigm shift in extreme compressed-matter physics?

"Only as experiment and tentative theory are together articulated to a match does the discovery emerge and the theory become a paradigm"

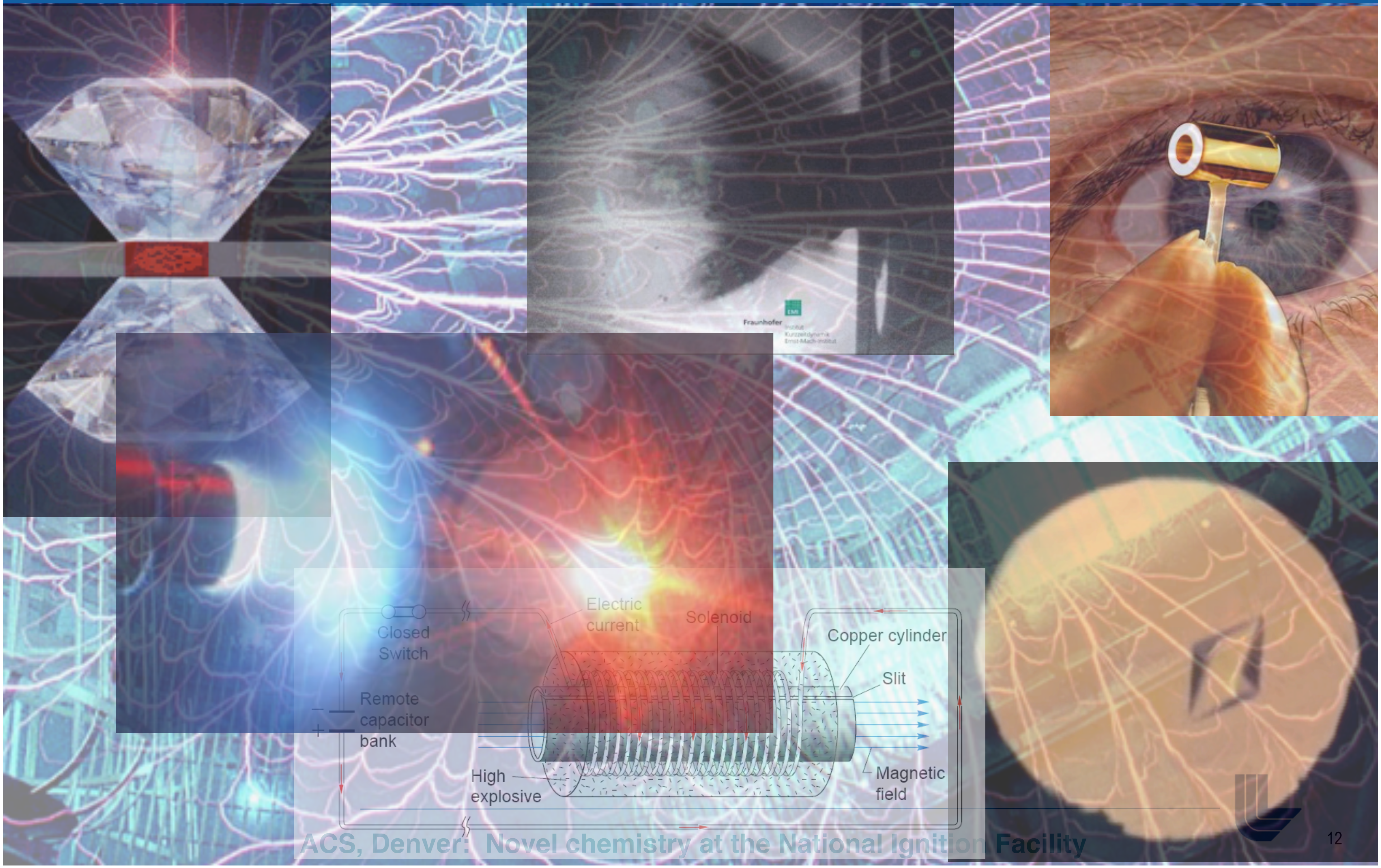
–p 61

"Further development ordinarily calls for the construction of elaborate equipment, the development of an esoteric vocabulary and skills, and a refinement of concepts. . .."

–p 64



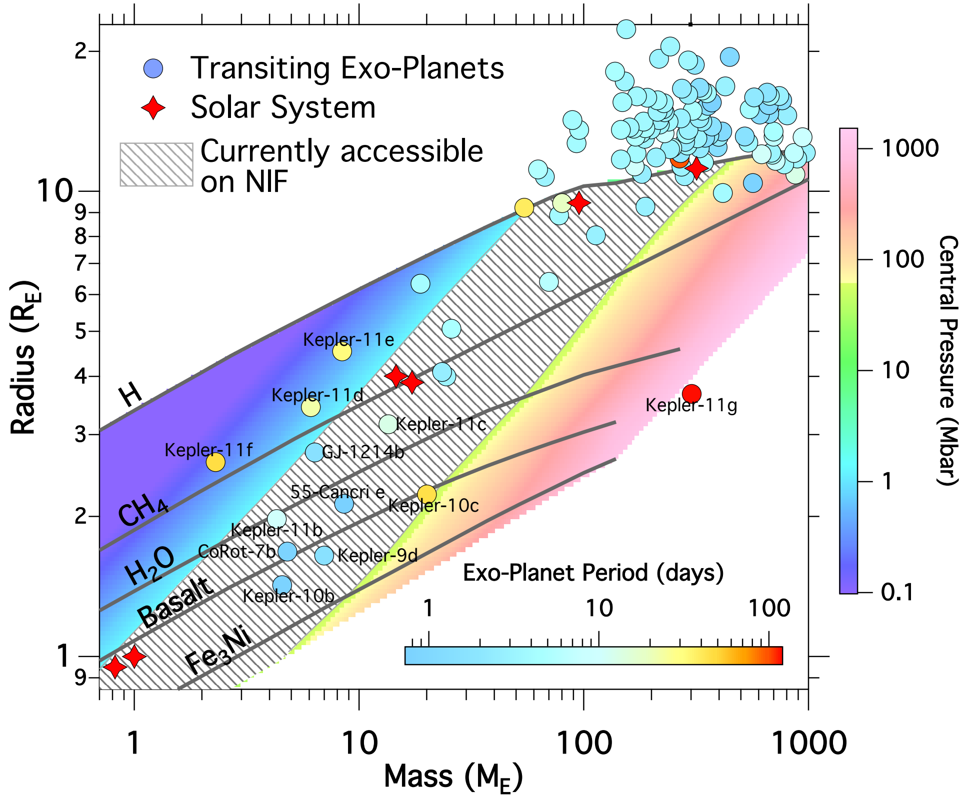
Where on earth do we have the "elaborate equipment" to study these emerging material states?

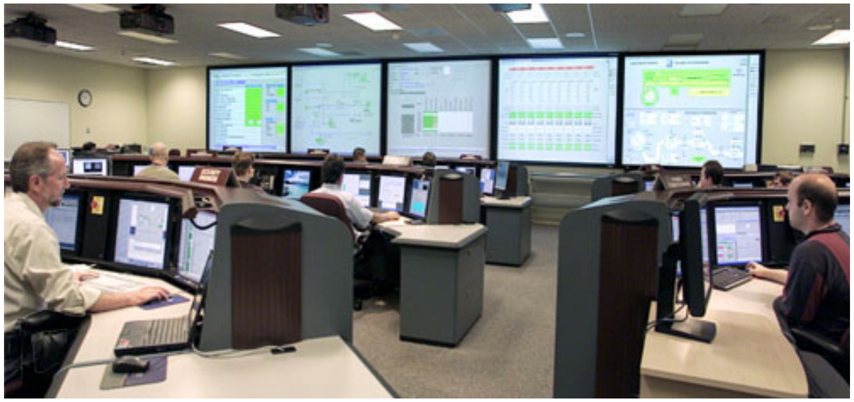



Only at the NIF
Developing a new paradigm for
Extreme Compression Science

NIF Ramp-Compression Experiments have already made the relevant exo-planet pressure range from 1 to 50 Mbar accessible.

This Spring we performed a series of experiments on the National Ignition Facilty



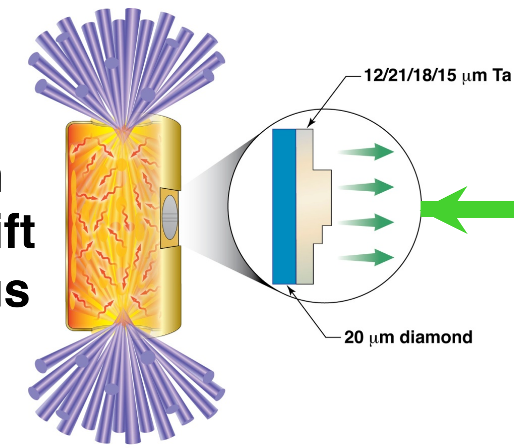


Our NIF experiments have demonstrated that we can access the relevant pressure composition region for exo-planet interiors.

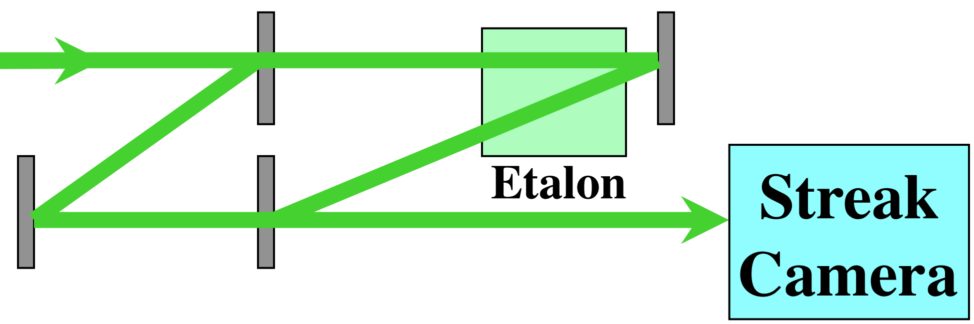
Laser-driven ramp-compression EOS experiments

VISAR—Velocity Interferometer System for Any Reflector Is our primary diagnostic

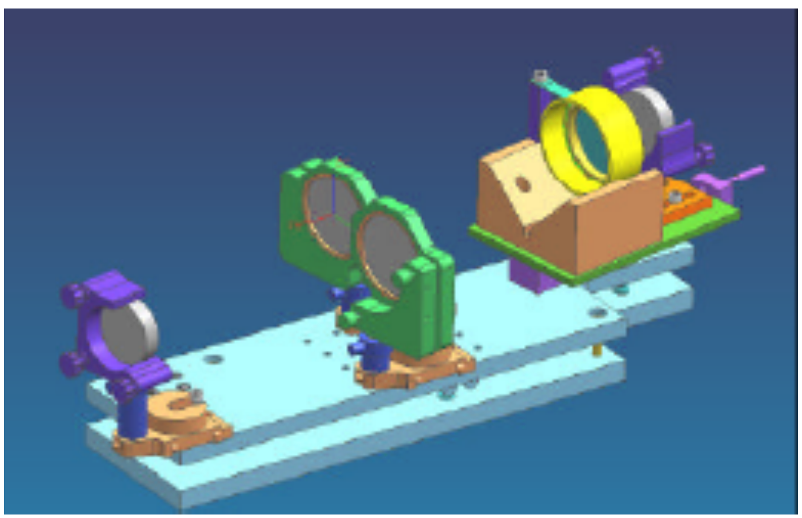
Imaging line-VISAR system gives phase shift (velocity) versus position

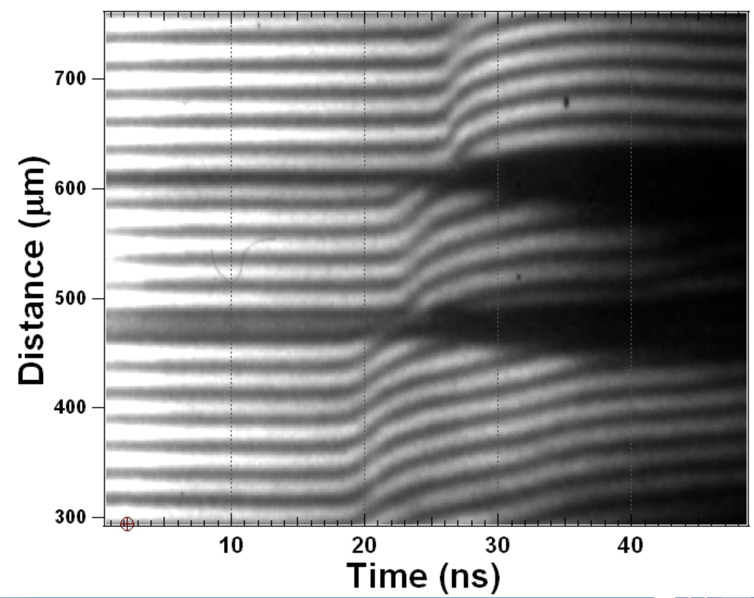


VISAR (Measures Doppler Shift)

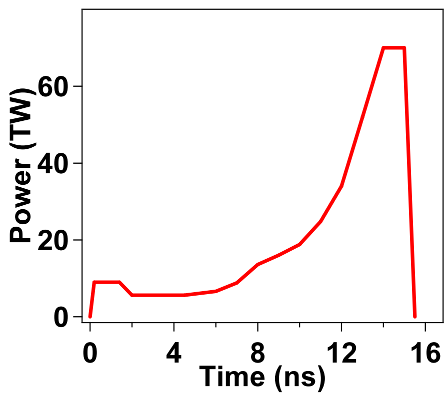


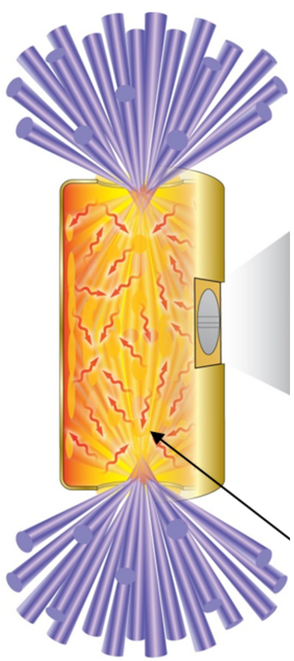
NIF-0310-18630

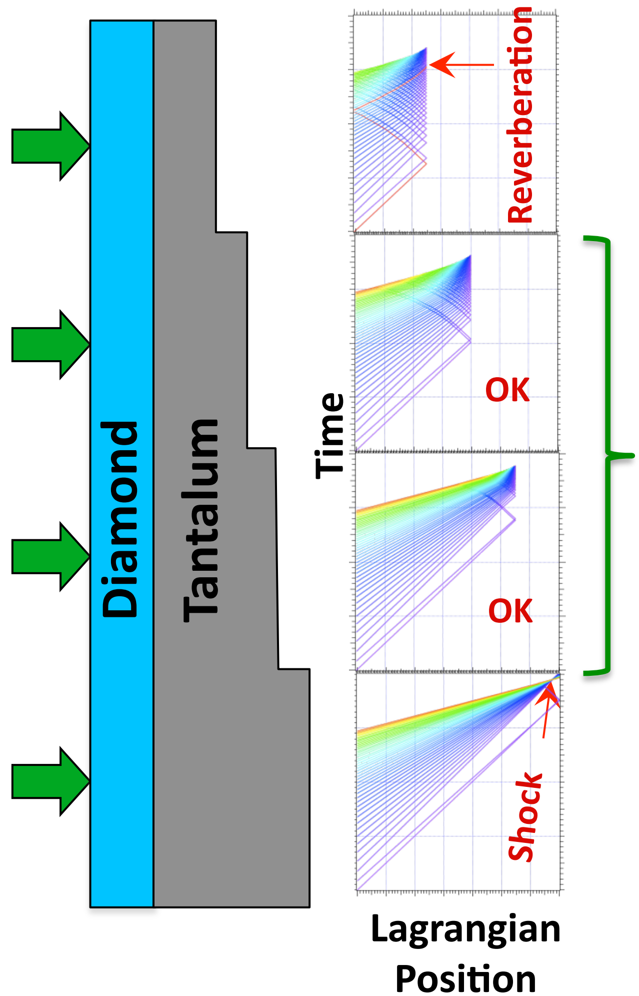


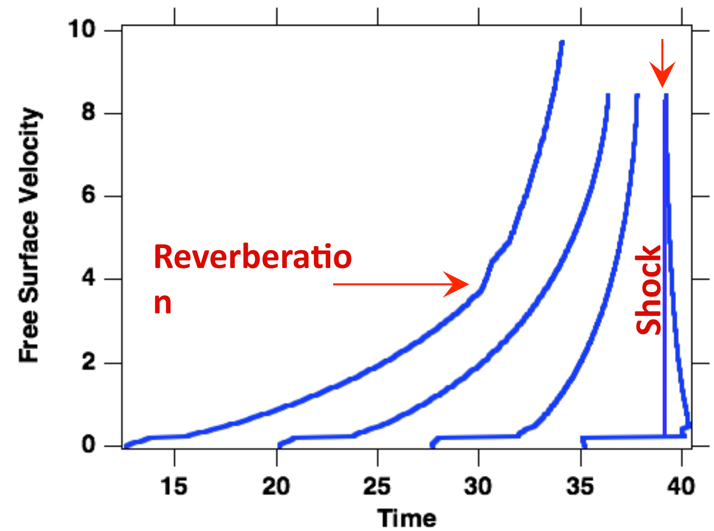


Under ramp loading, EOS (stress-density) can be determined from free-surface velocity measurements









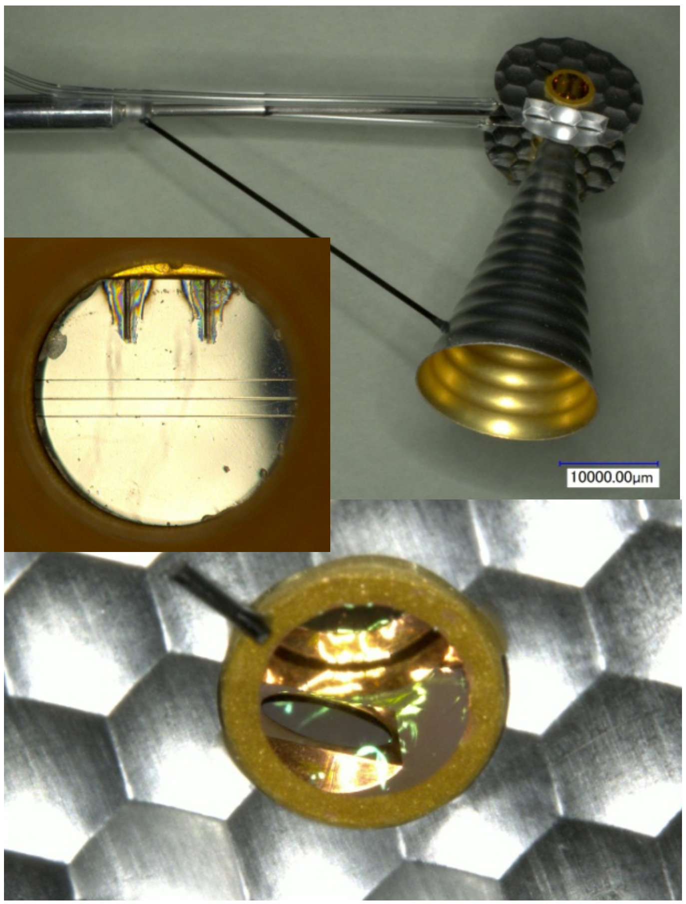
Design constraints:

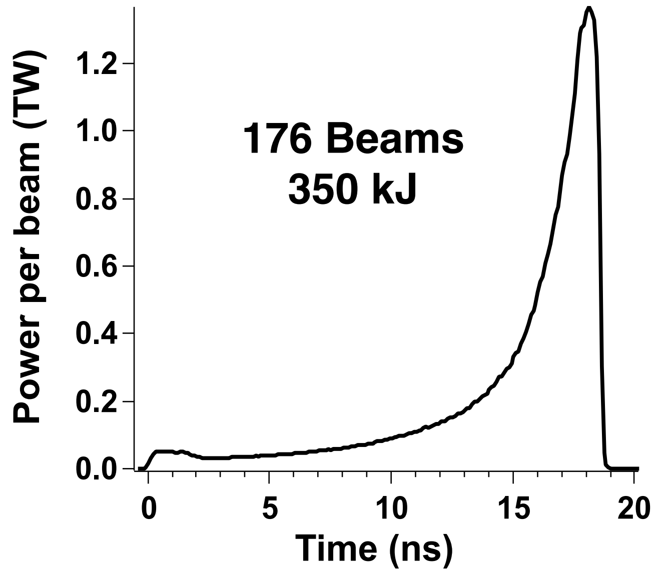
No reverberation <u>and</u> no shock

Nano-crystalline diamond EOS on NIF

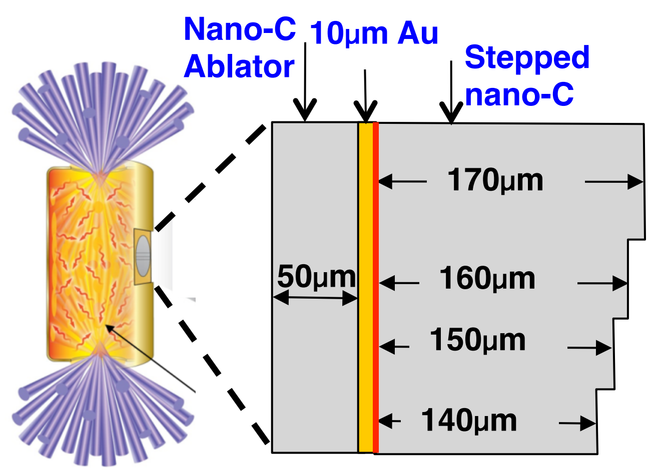
We studied the stress-density of solid diamond on the NIF

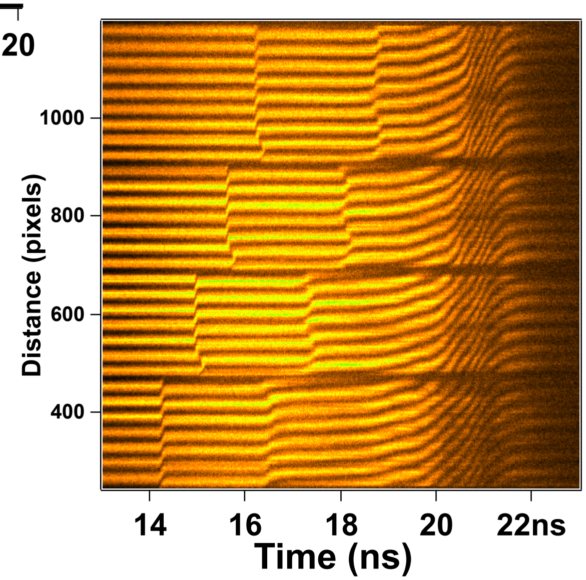
Reseachers: Ray Smith, Jon Eggert, Dave Braun (LLNL), Raymond Jeanloz (UCB), Tom Duffy and Jue Wang (Princeton)





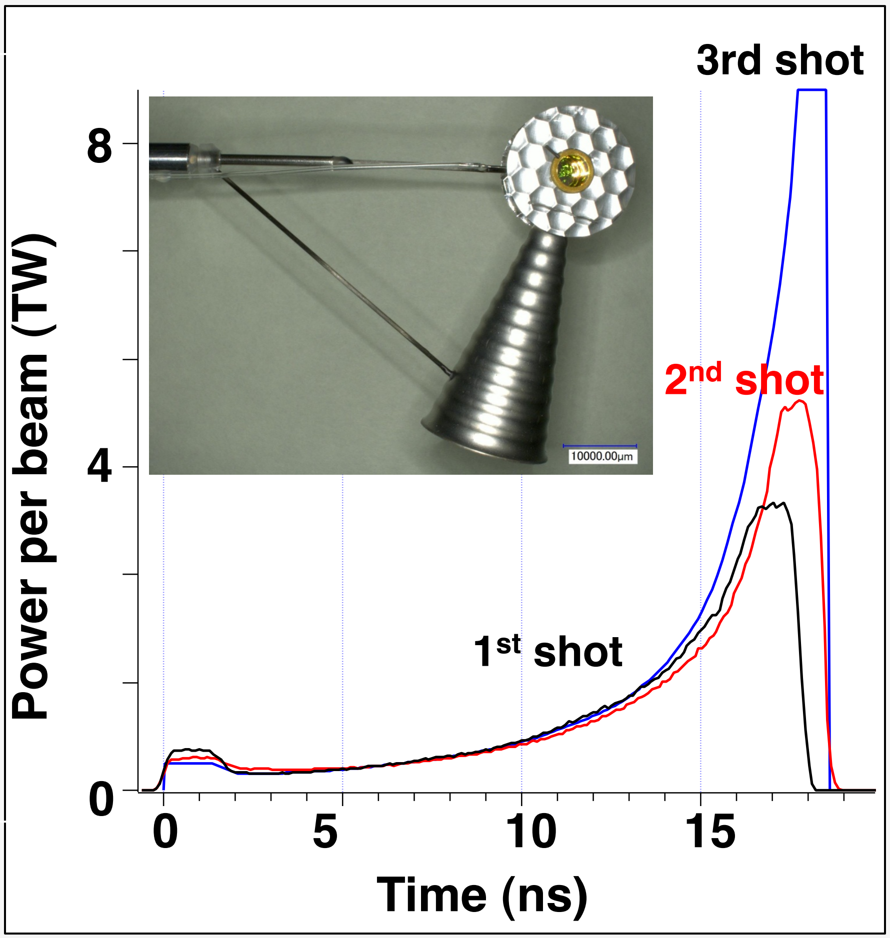
Exquisite control of the laser pulse shape is critical to success



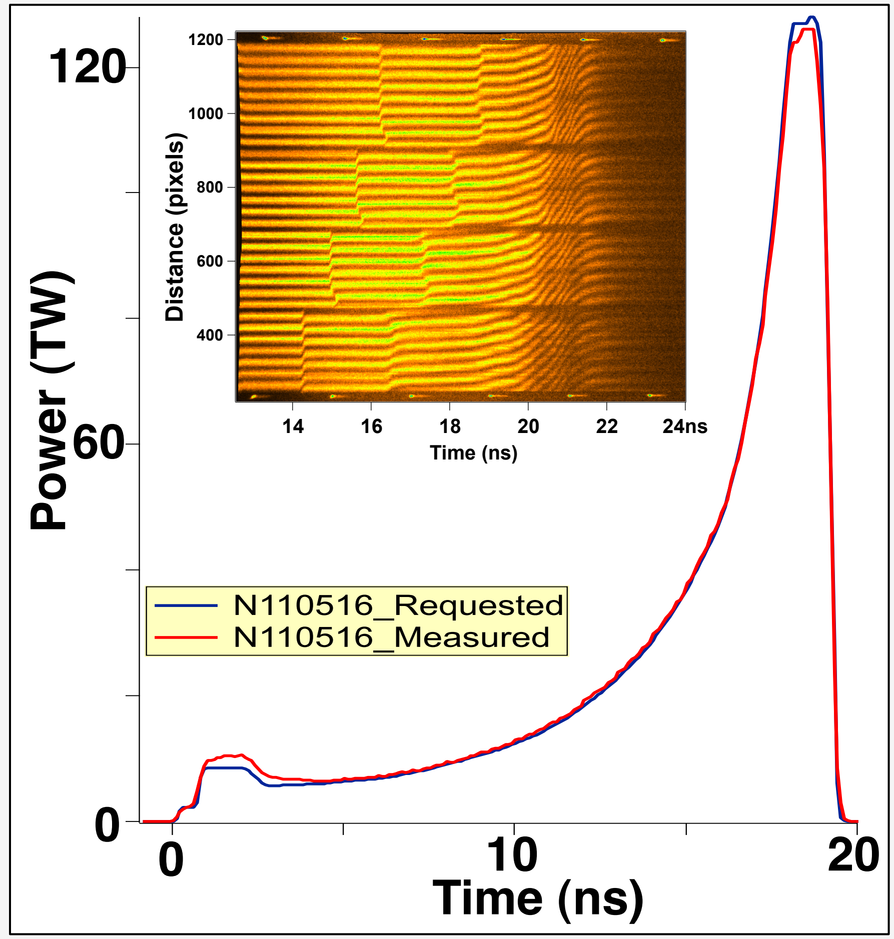


In addition to the higher energy afforded by NIF, pulse shaping was key to achieving higher pressure

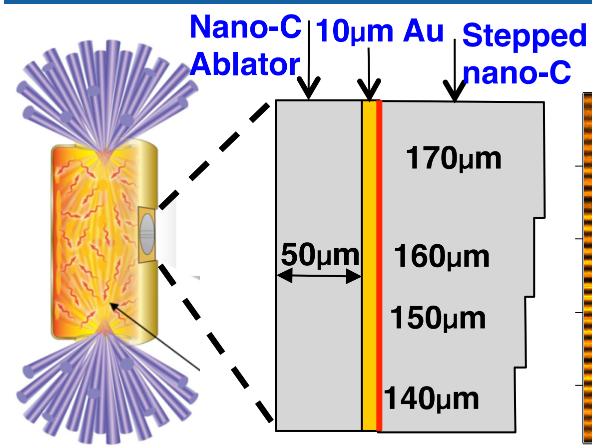
Pulse Shape evolution for diamond ramp experiments

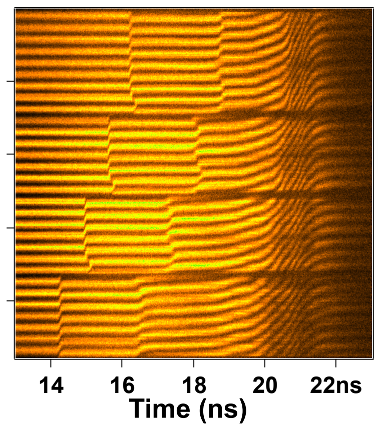


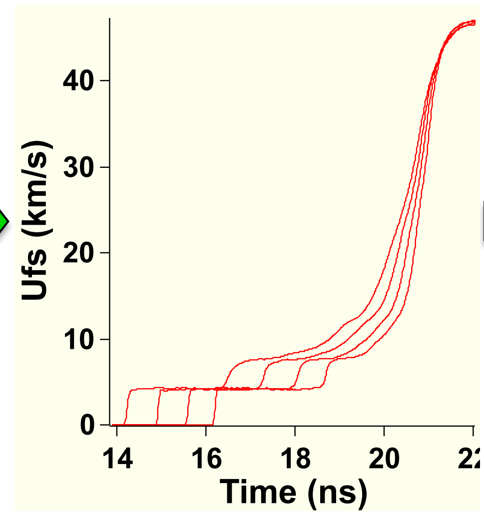
Requested vs delivered pulse shapes for ramp experiments



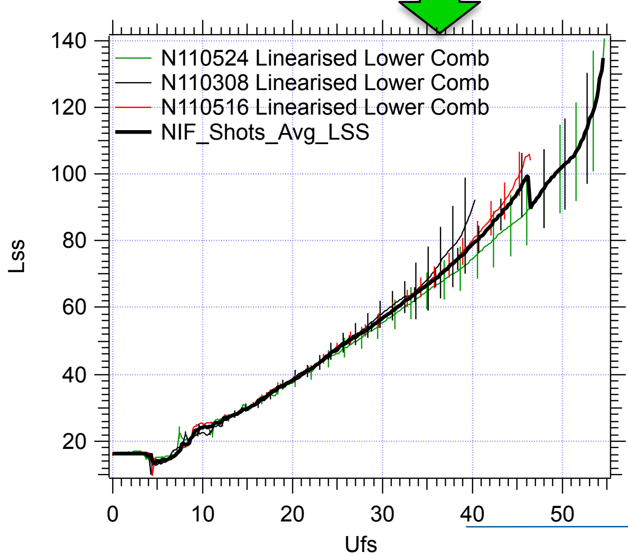
We use an Iterative Lagrangian Analysis to extract stress and density (Rothman, et al., (2005)







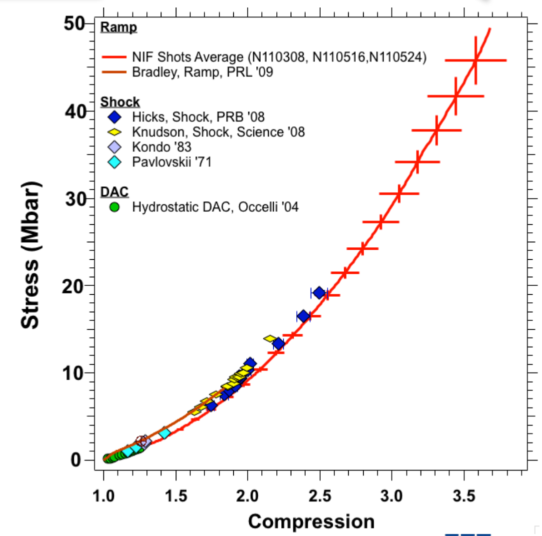
$$C_L(u) = \frac{\Delta x}{\Delta t}$$



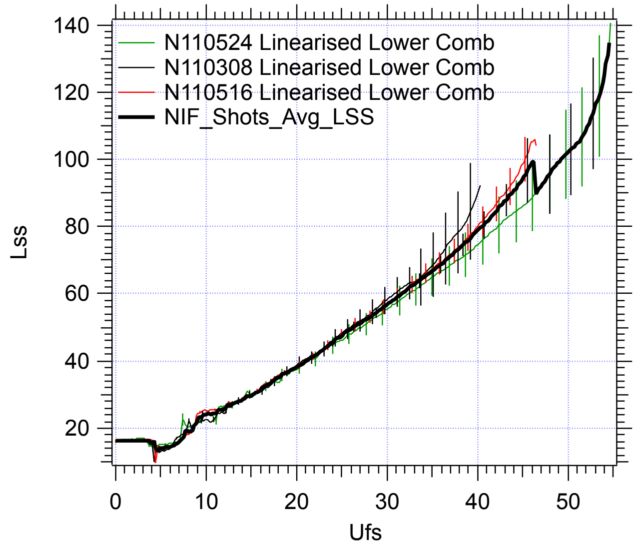
Since we measure free surface velocity, not *u* we must use an iterative correction developed by Rothman (2005)

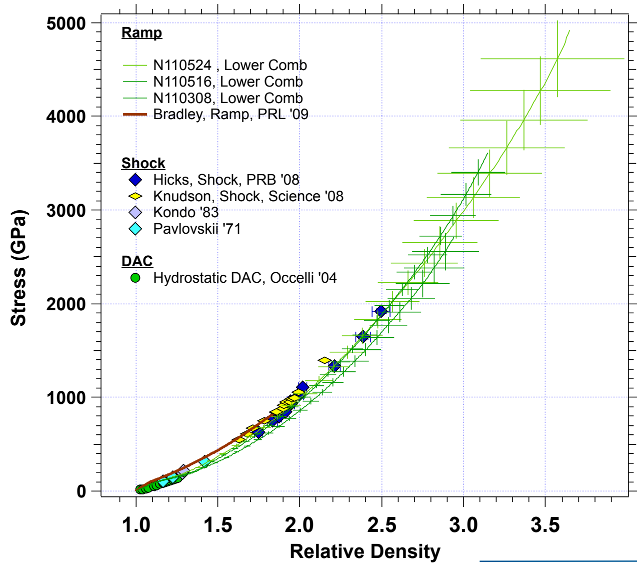
$$\rho(u) = \rho_0\left(1 - \int du/C_L\right)^{-1}$$

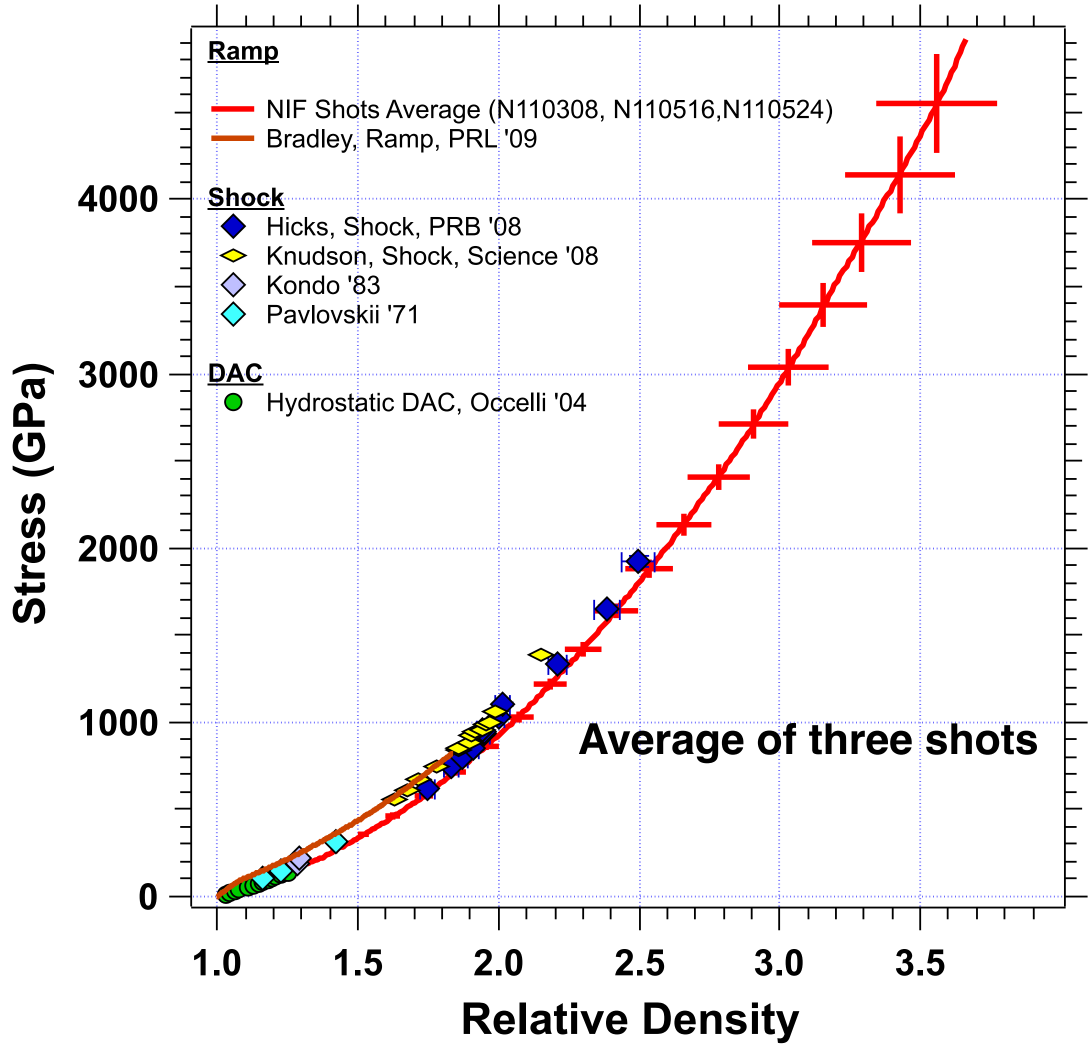
$$P_x(u) = \rho_0 \int C_L \, du$$



Ramp-compression EOS of nano-crystalline diamond to 50 Mbar.

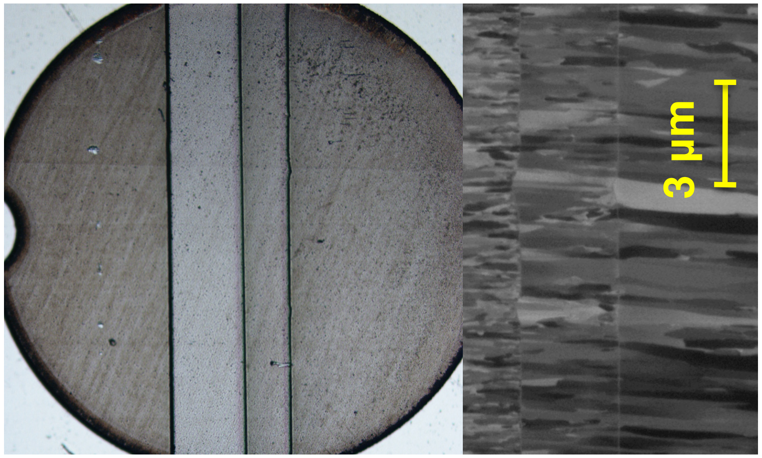


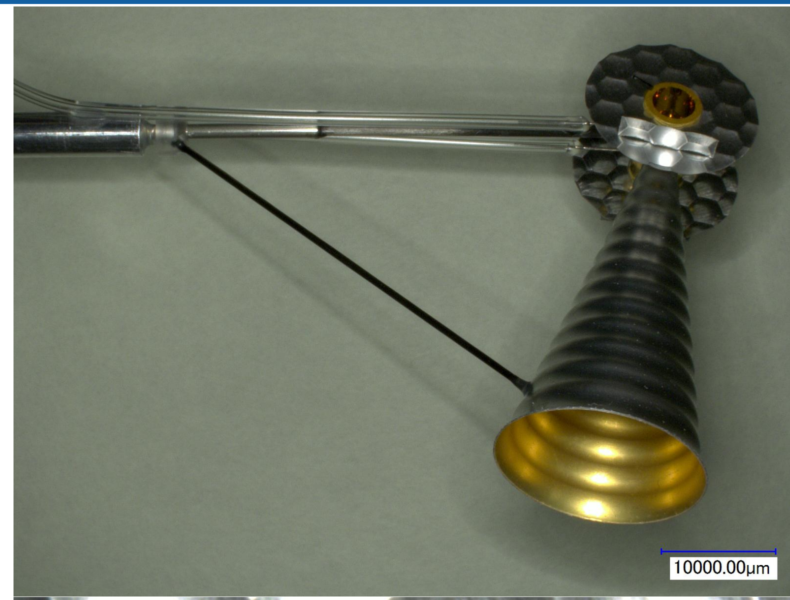


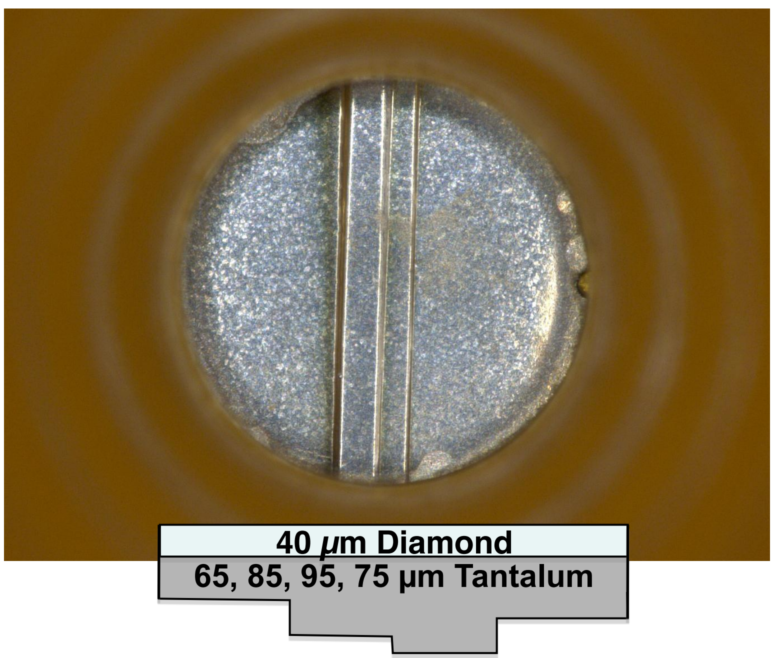


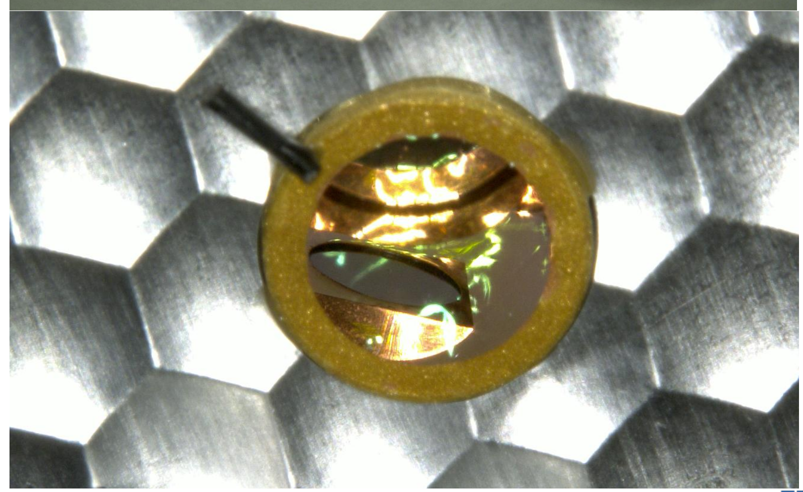
Vapor-deposited tantalum EOS on NIF

NIF: Vapor-deposited tantalum targets

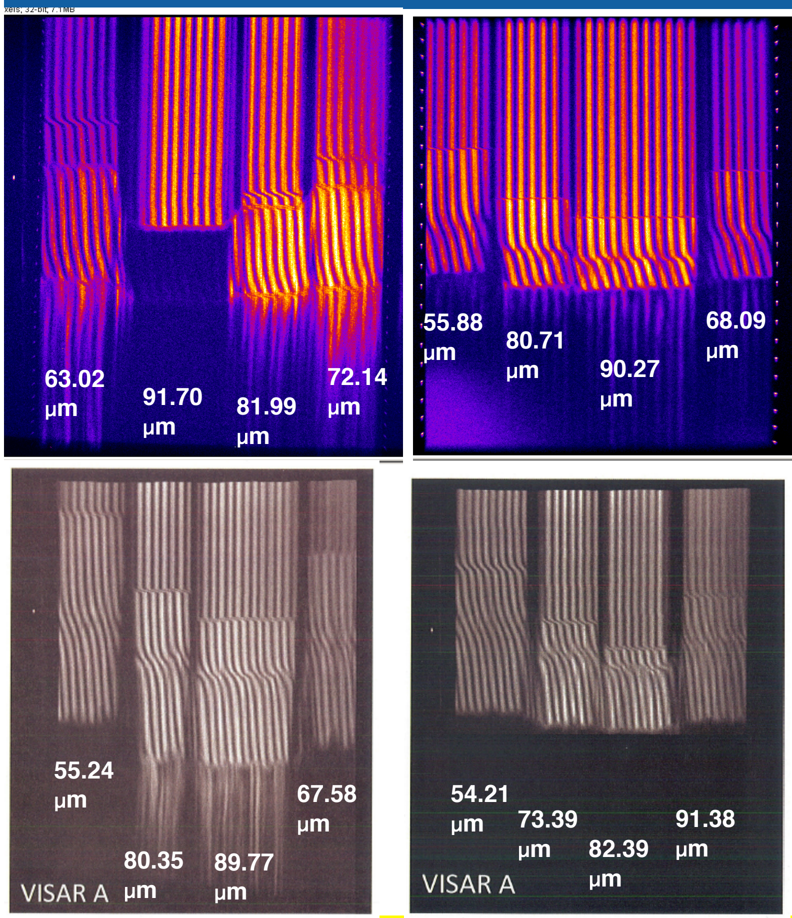






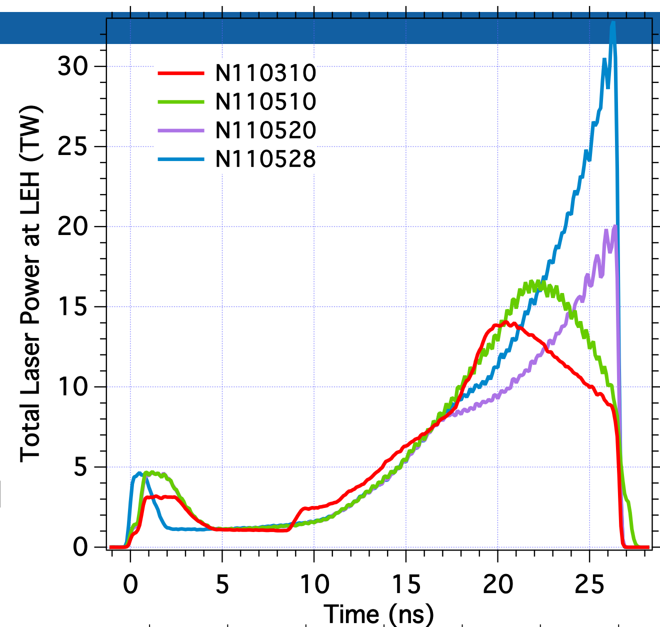


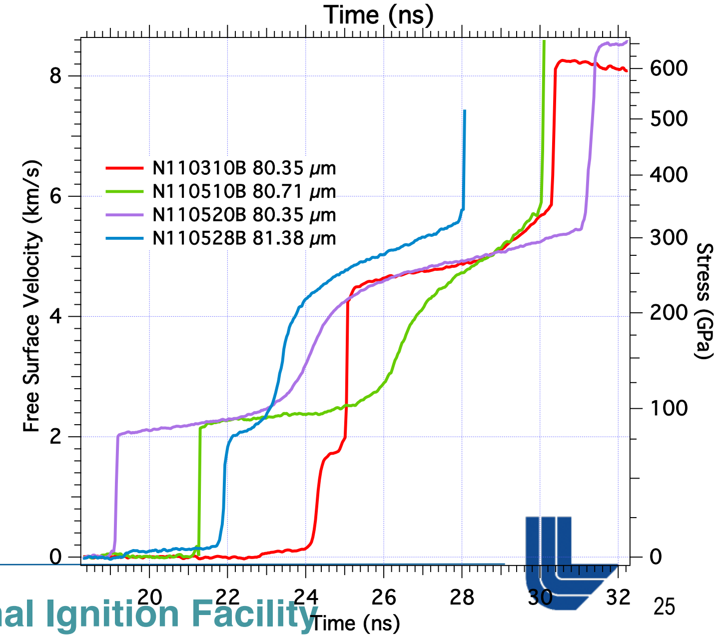
Unlike diamond shock initiation begins in tantalum at 3.4 Mbar, independent of pressure-drive



Variation of pressure drive (stress rate) is much larger than was needed for diamond.

Repeated shock initiation at ~ 3.4 Mbar with only minor dependence on sample thickness and drive profile.

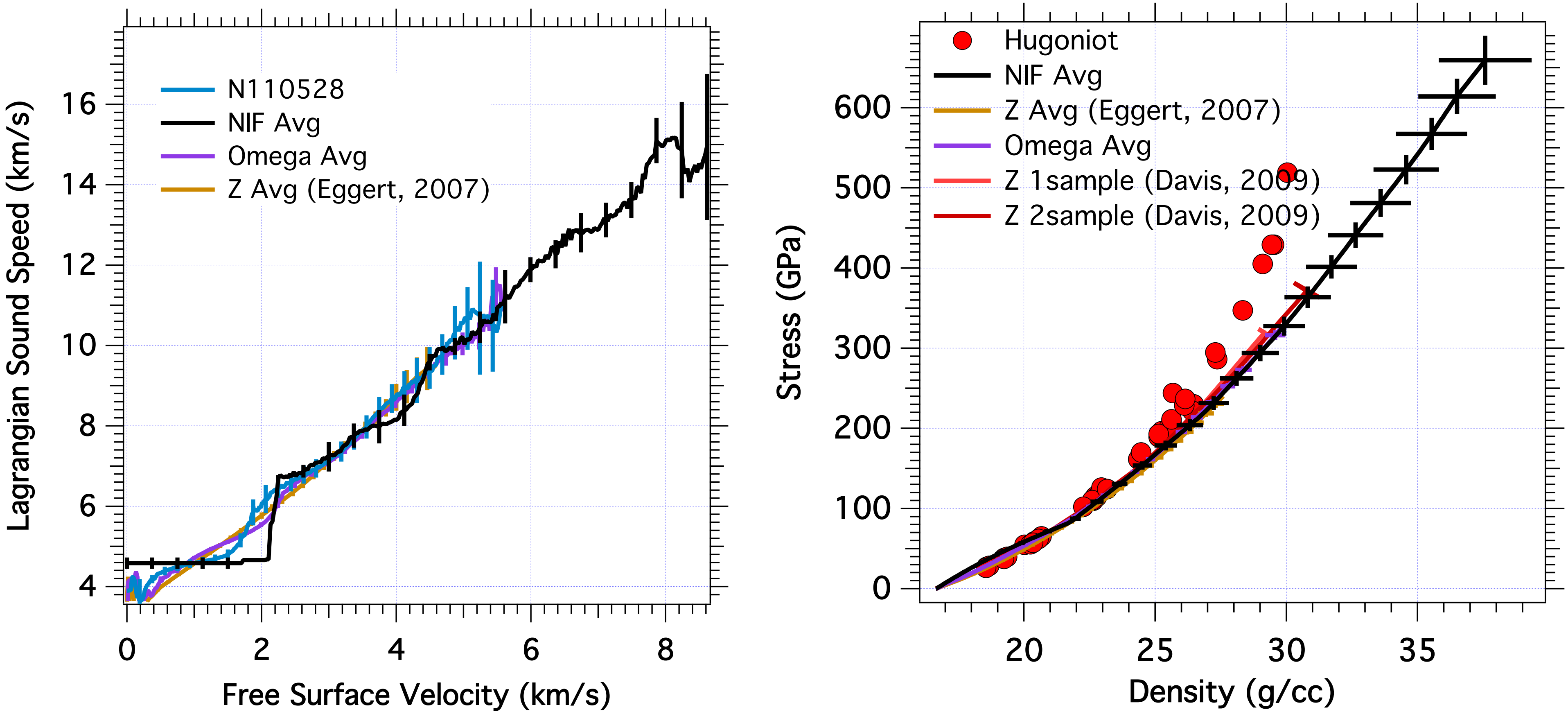




These results strongly suggest a constitutive (e.g. phase transition) rather than a hydrodynamic driver for the shock.

Even with the shock, we extract the sound speed and stress-density for multiple shots

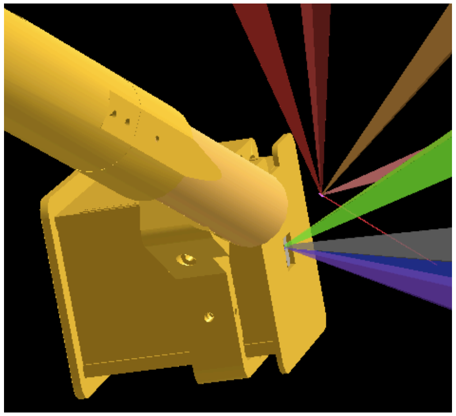
Excellent agreement with previous data from Z and Omega

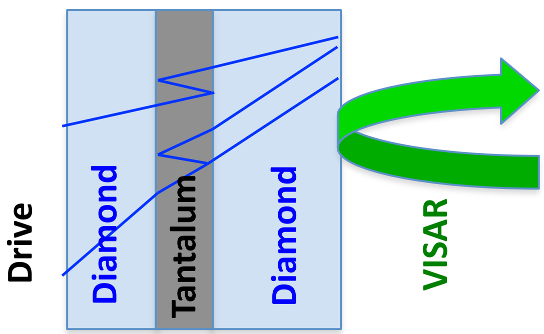


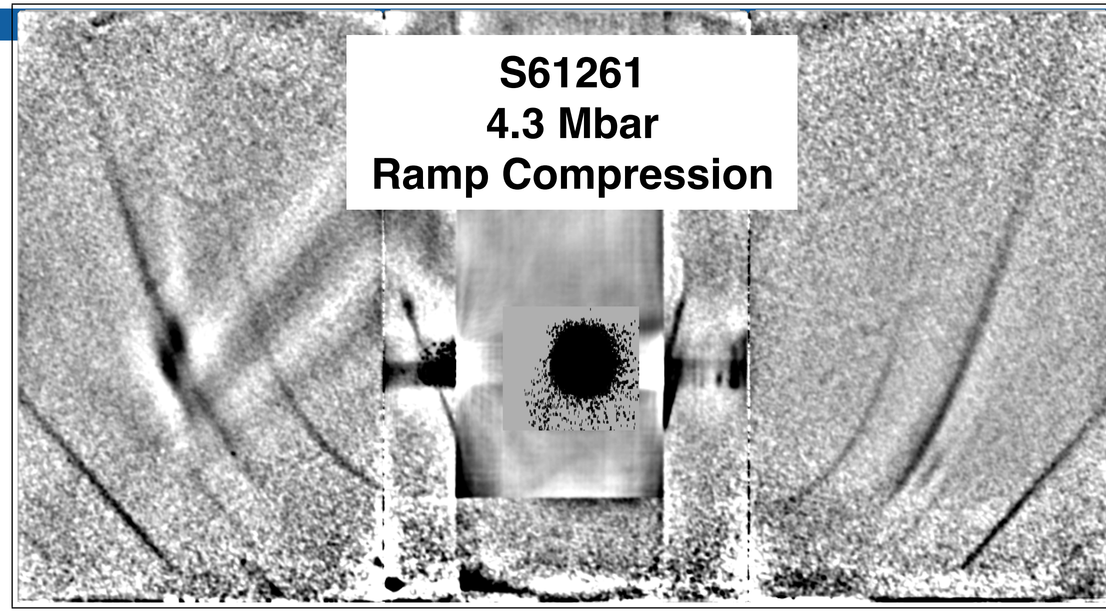
These experiments deliver important information about the stress-strain relation of materials, but we also need more direct measurements.

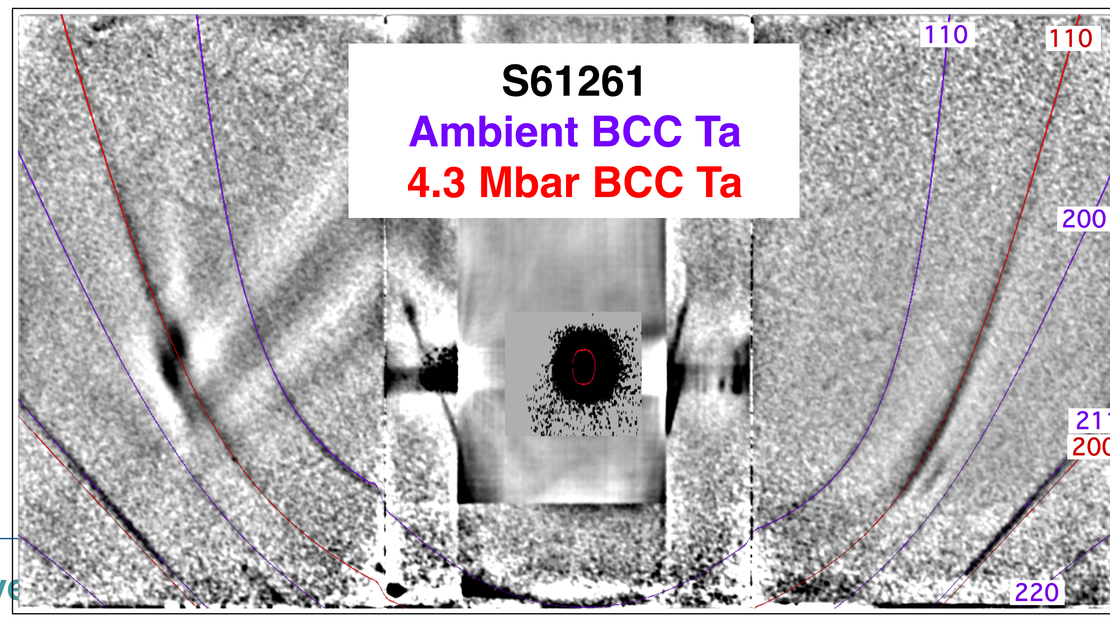
Powder x-ray diffraction of rolled tantalum on the Omega laser

We performed high-pressure x-ray diffraction on tantalum at the Omega laser







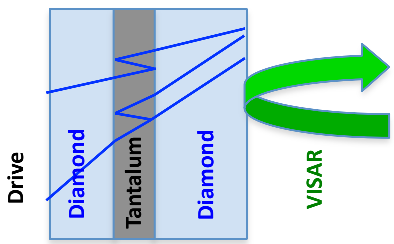


Diffraction data quality is roughly where DAC diffraction was in the '80s. We need to make similar strides.

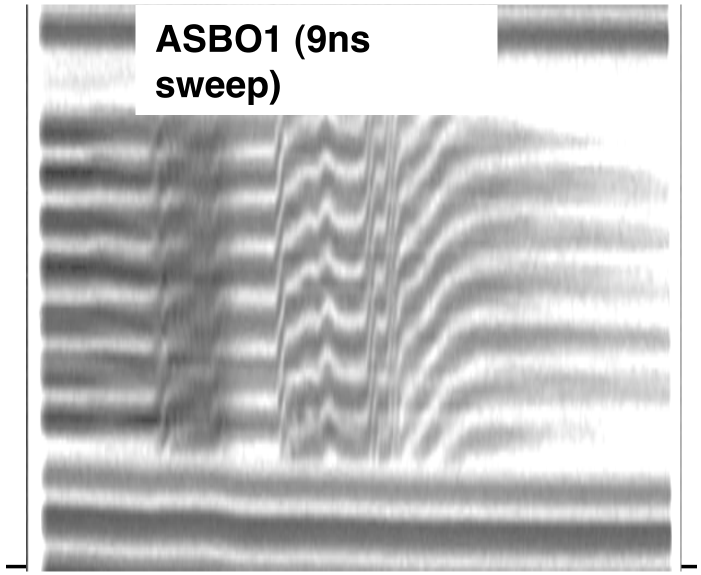
We determine stress by backward integration of diamond free-surface velocity

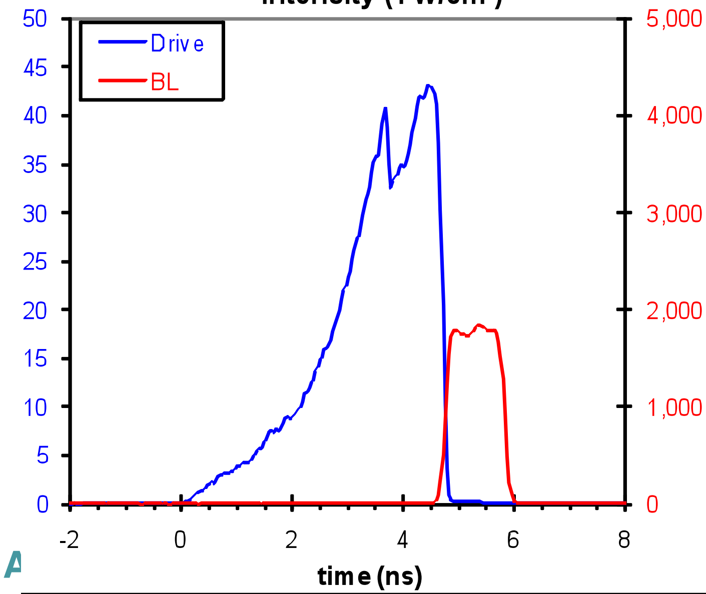
Shot 61261,
OMEGA 2011-0223

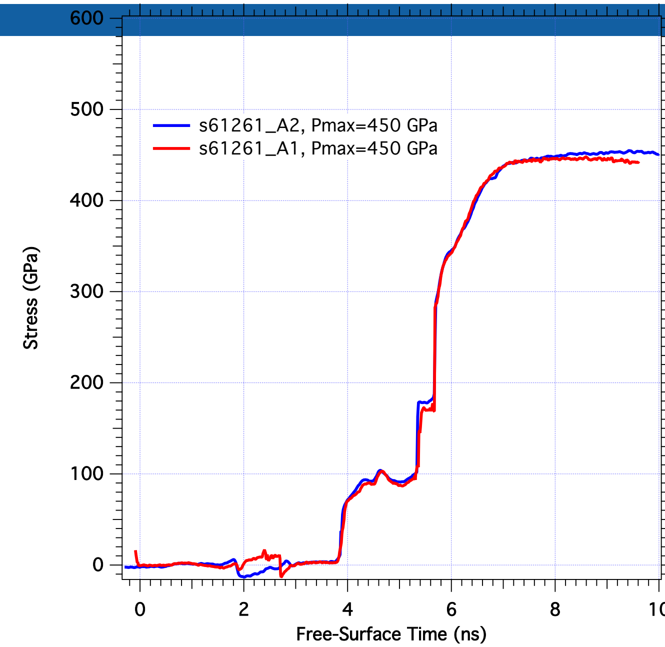
Target: C[17]Ta[3]C [40], BL: Fe

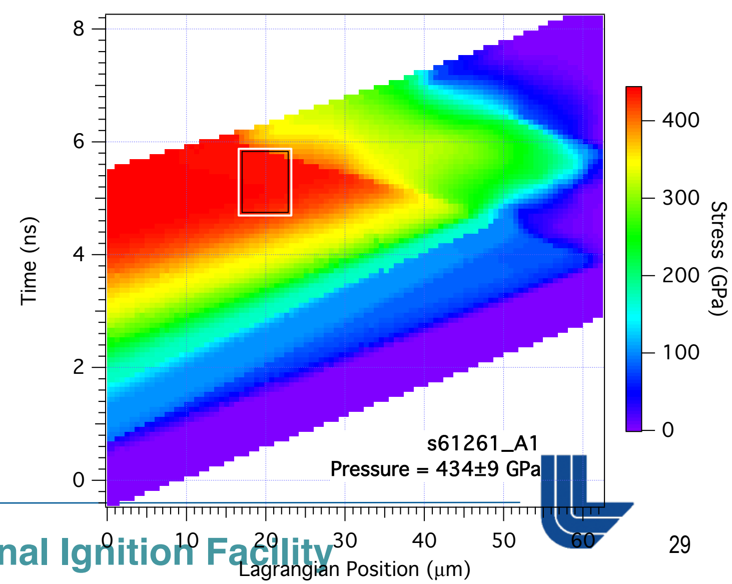


Ramp drive: 246 J
(t_{BL} = 4.6 ns)
P = 4.34±0.09 Mbar

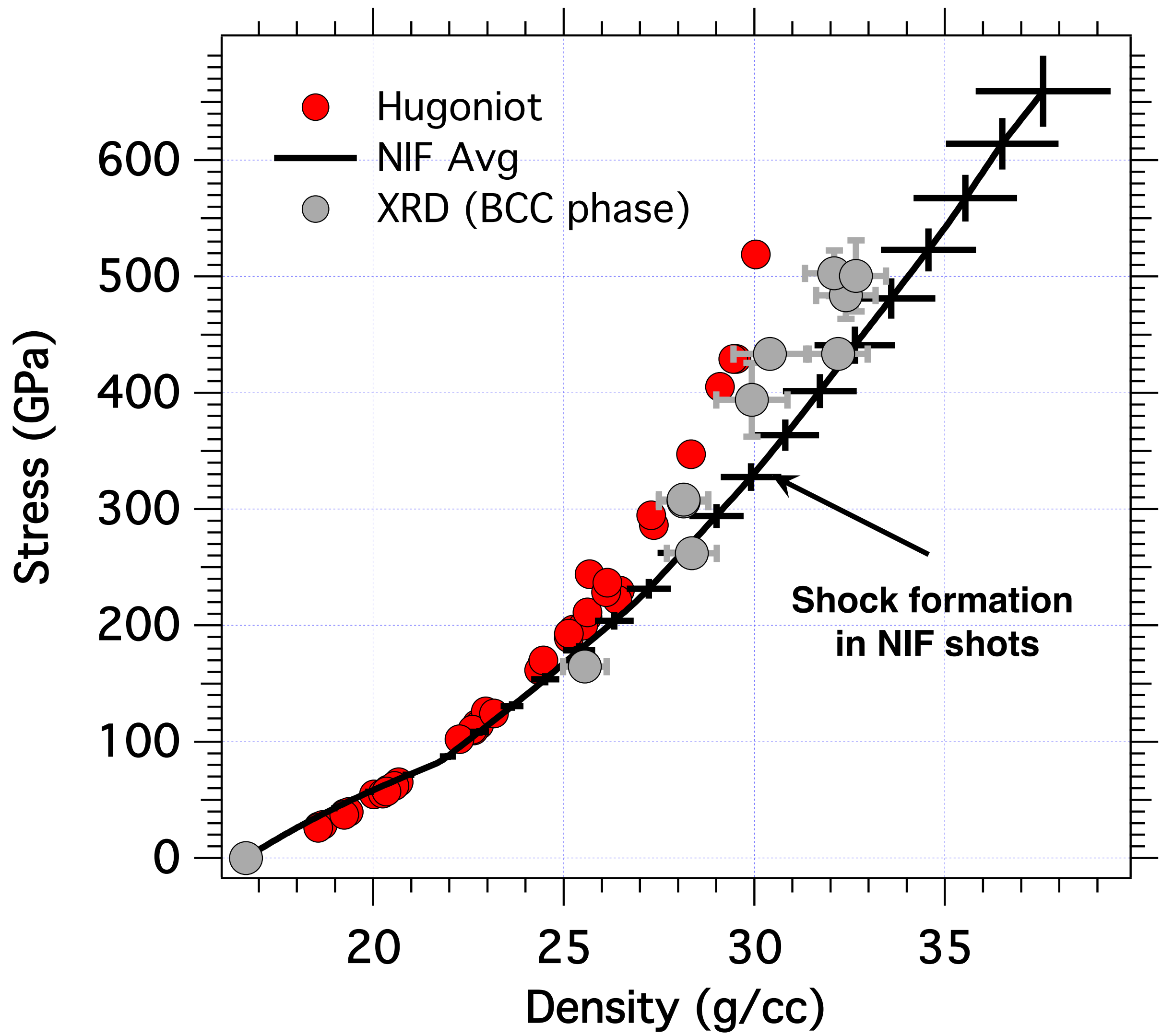




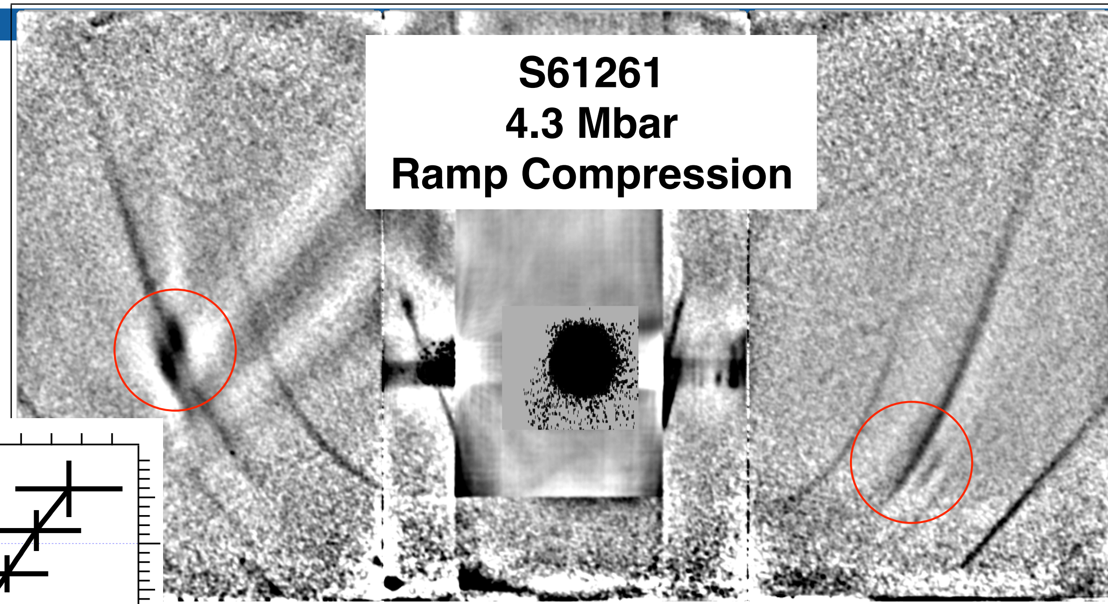


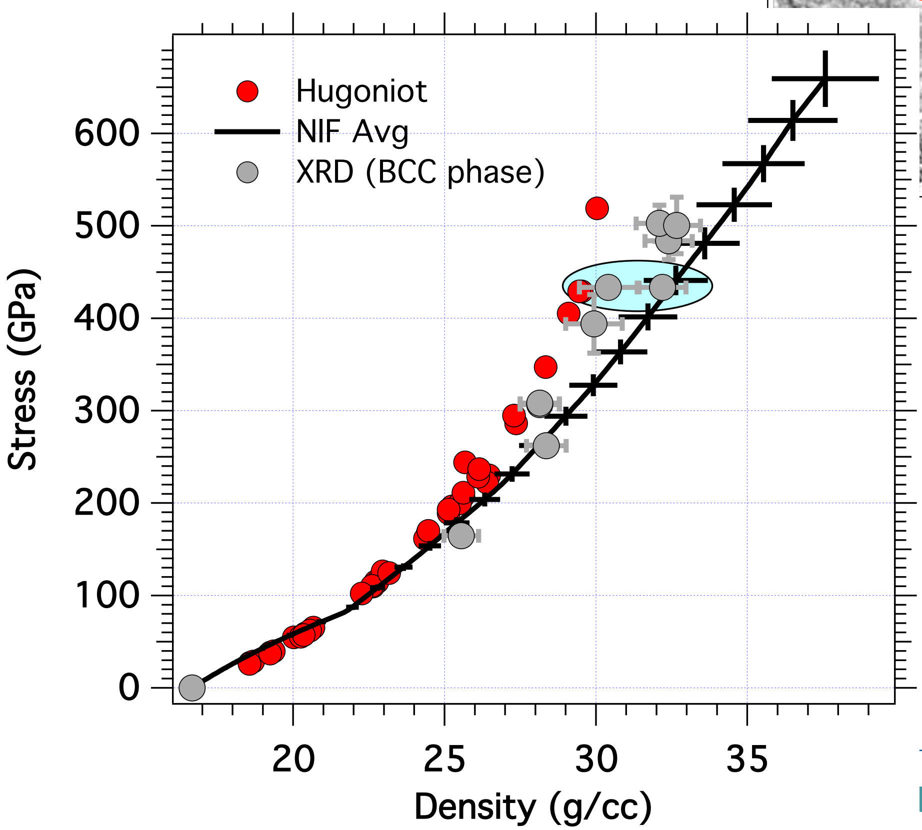


We have measured the (assumed) BCC density for 8 shots.



Diffraction for S61261 can be interpreted as two phase coexistence

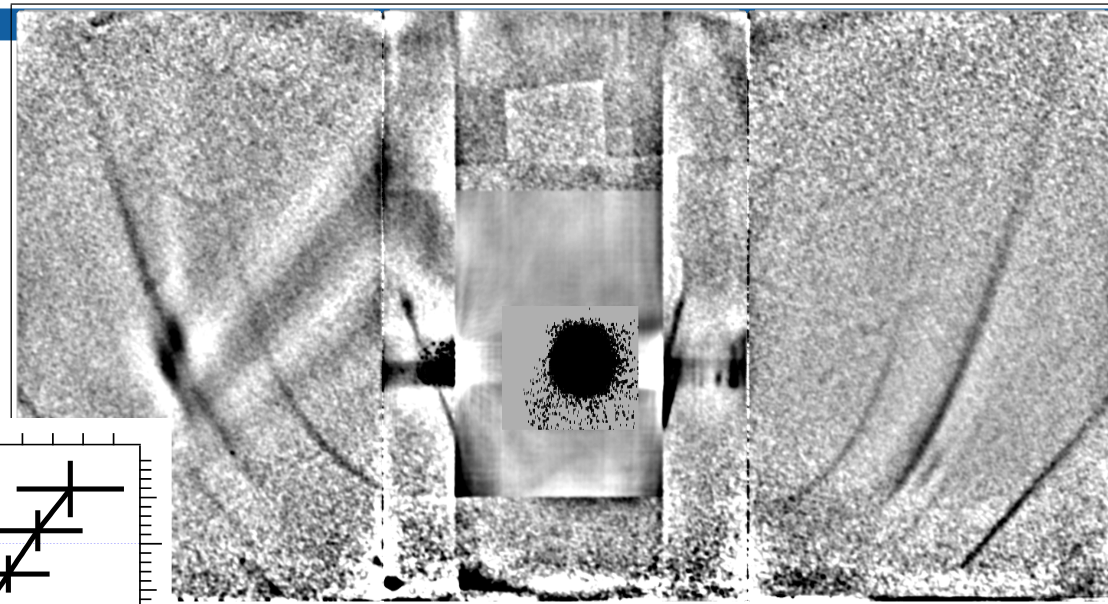


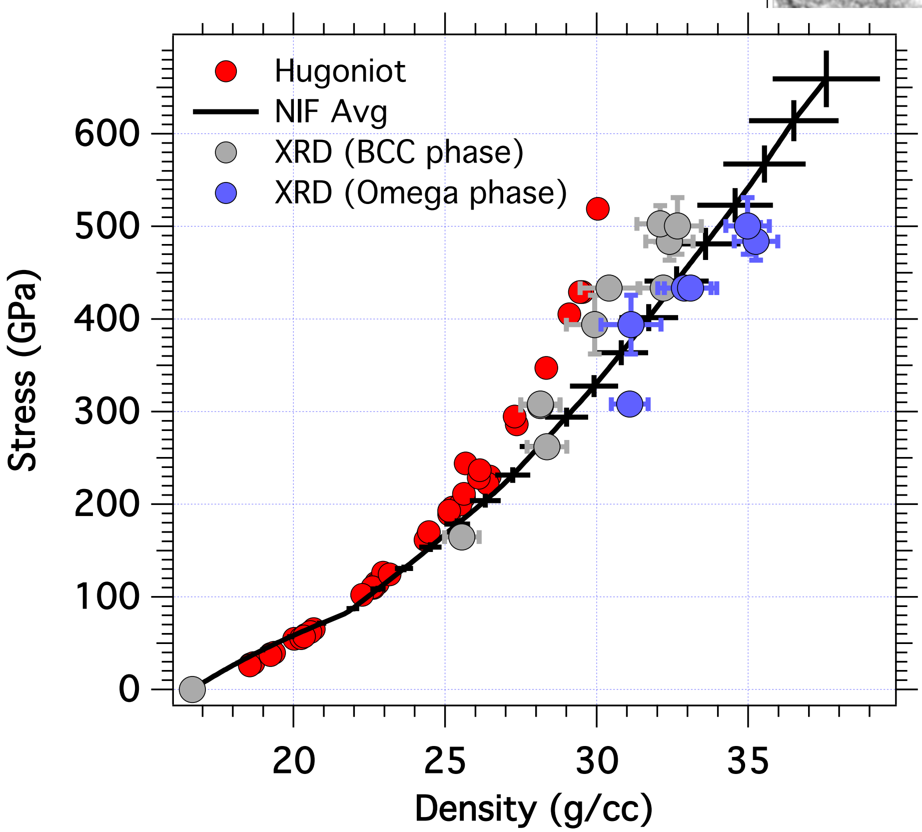


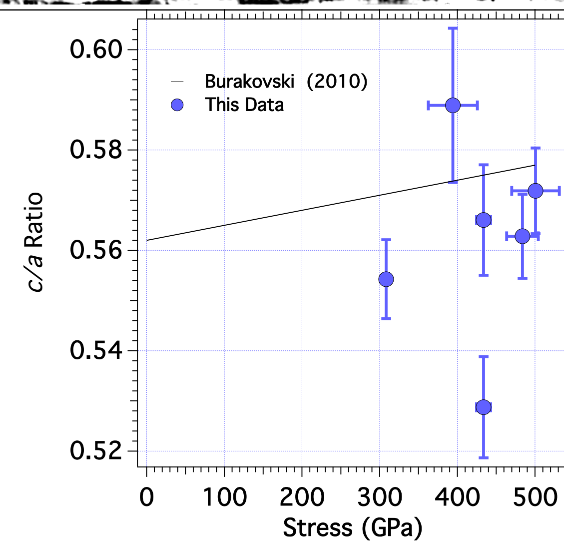
A consistent understanding of both the NIF EOS and the Omega diffraction data can be had by positing a Ta phase transition near 3.4 Mbar.

e.g. Burakovski (2010) predict ω-phase.

We can also determine density assuming the omega phase

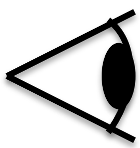




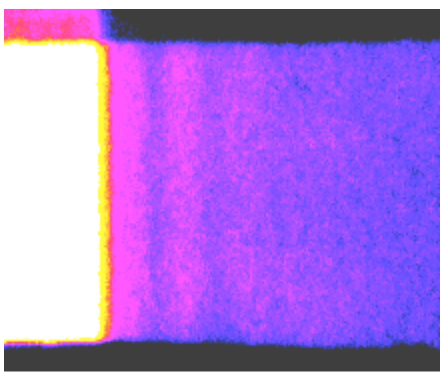


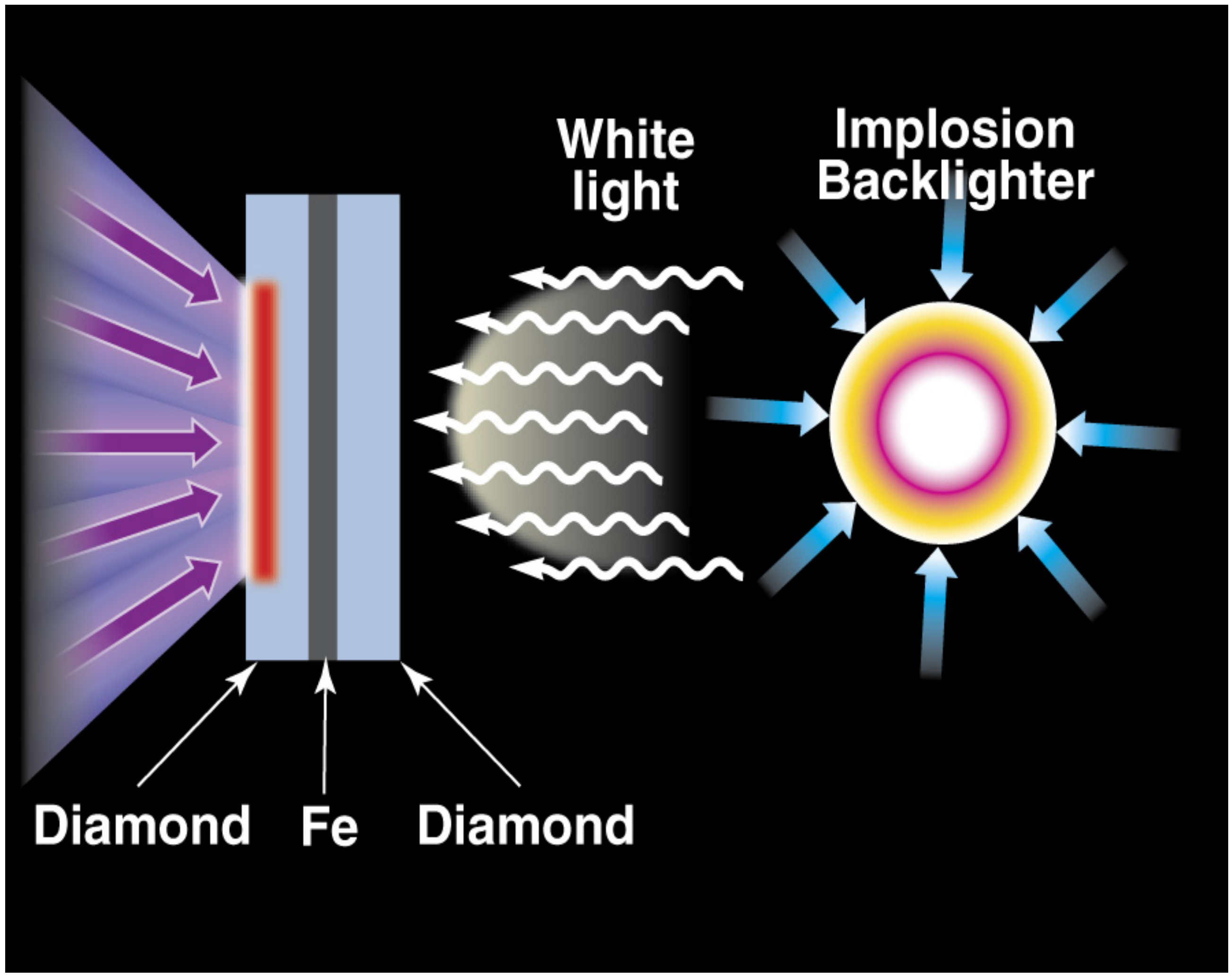
Extended X-ray Absorption Fine Structure (EXAFS) at the Omega Laser

Experimental design for EXAFS is very similar to difraction

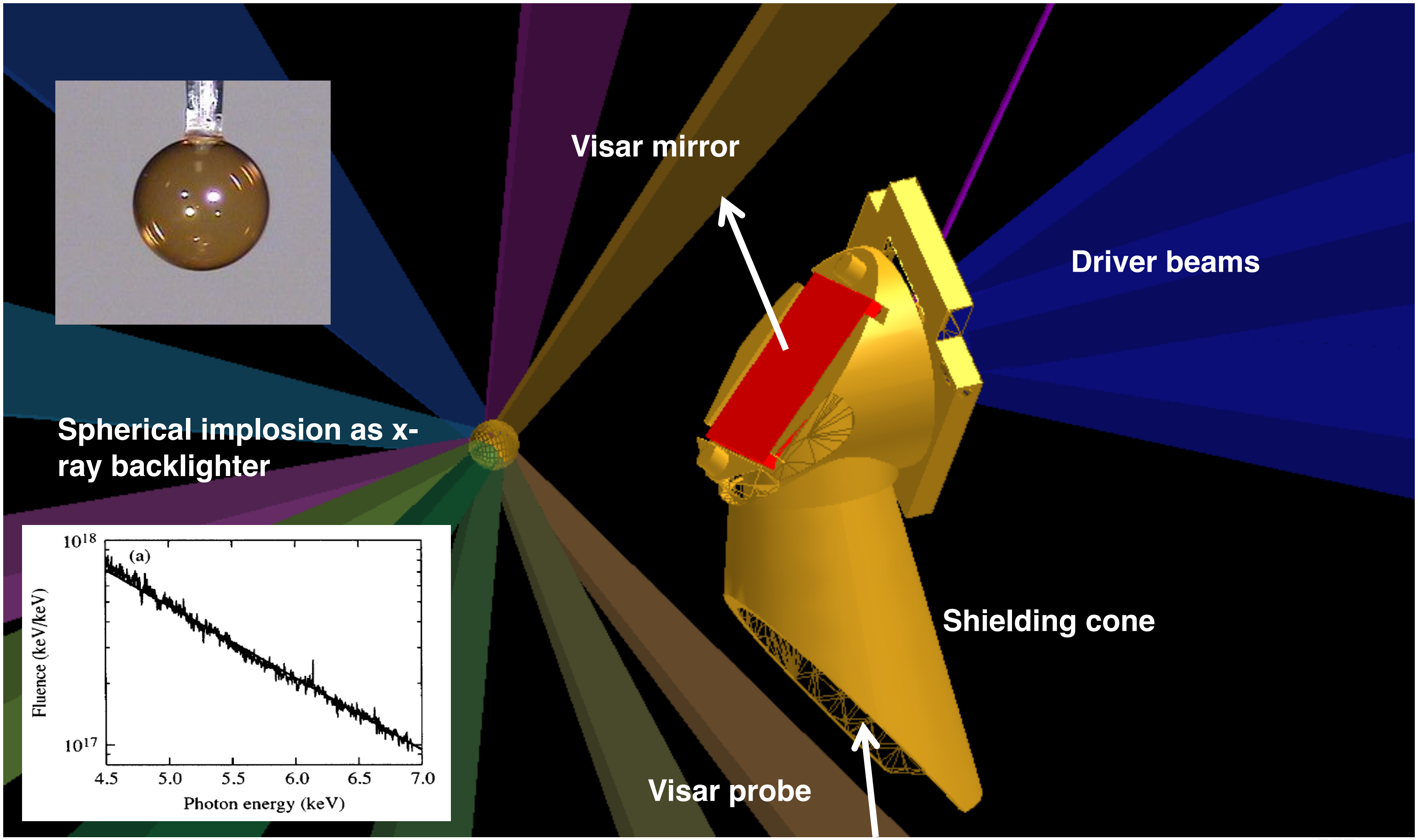


X-ray absorption
spectrum

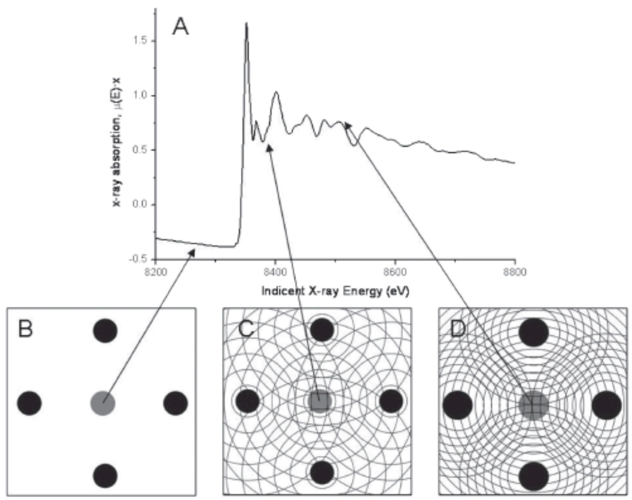




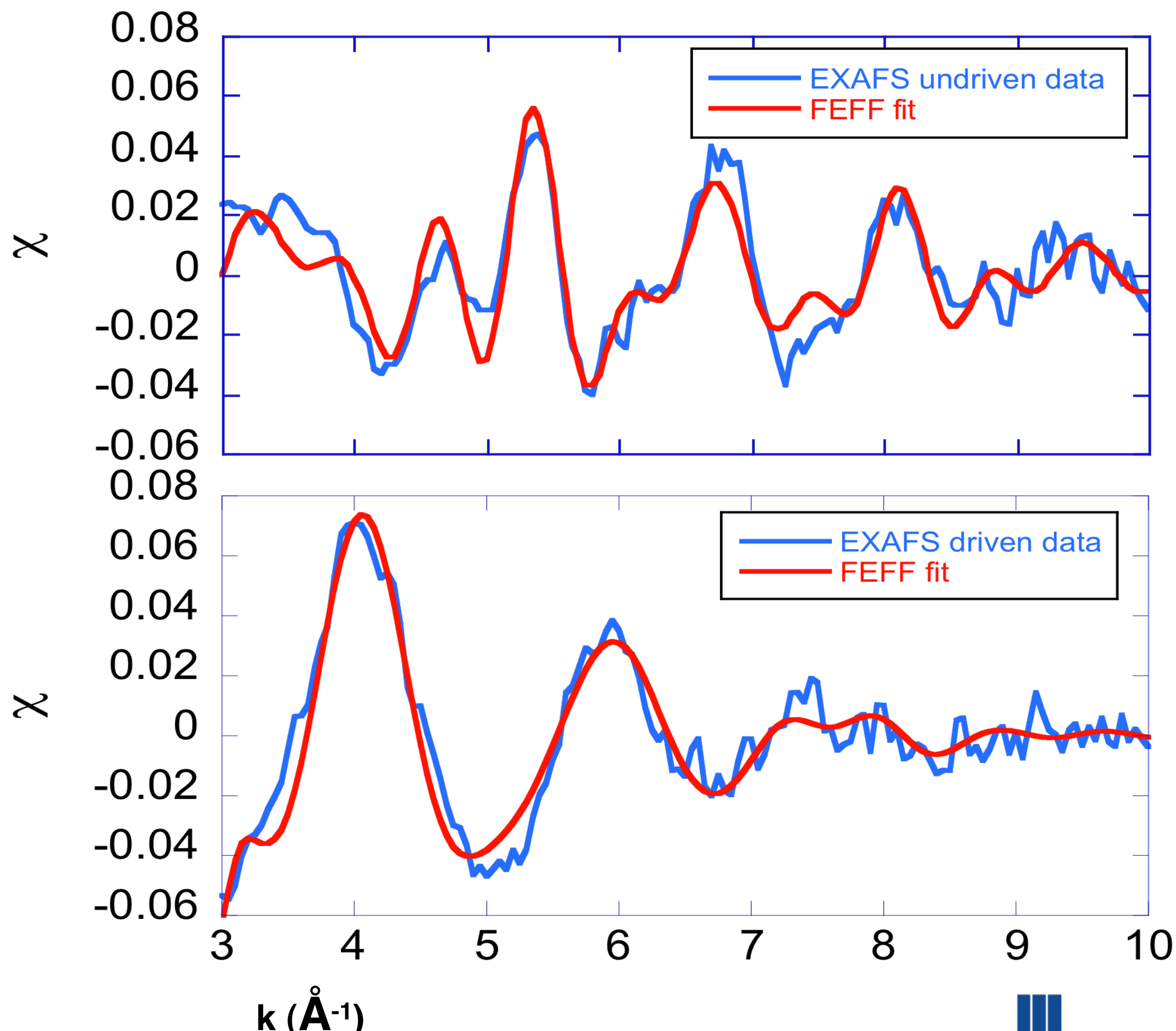
Omega experimental setup for EXAFS



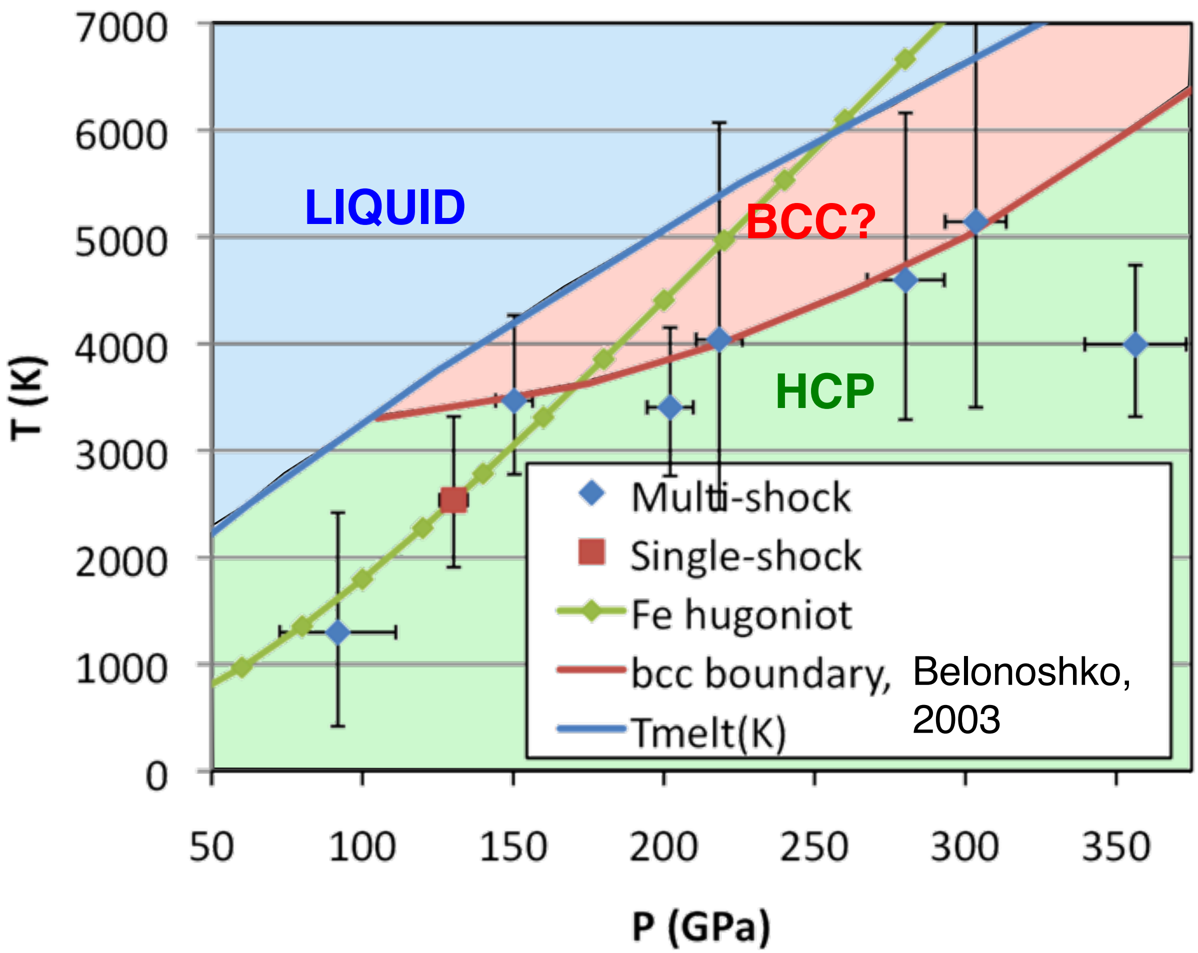
EXAFS data is sensitive to short-range structure, density, and temperature



EXAFS is sensitive to temperature through the Debye-Waller factor (DWF)



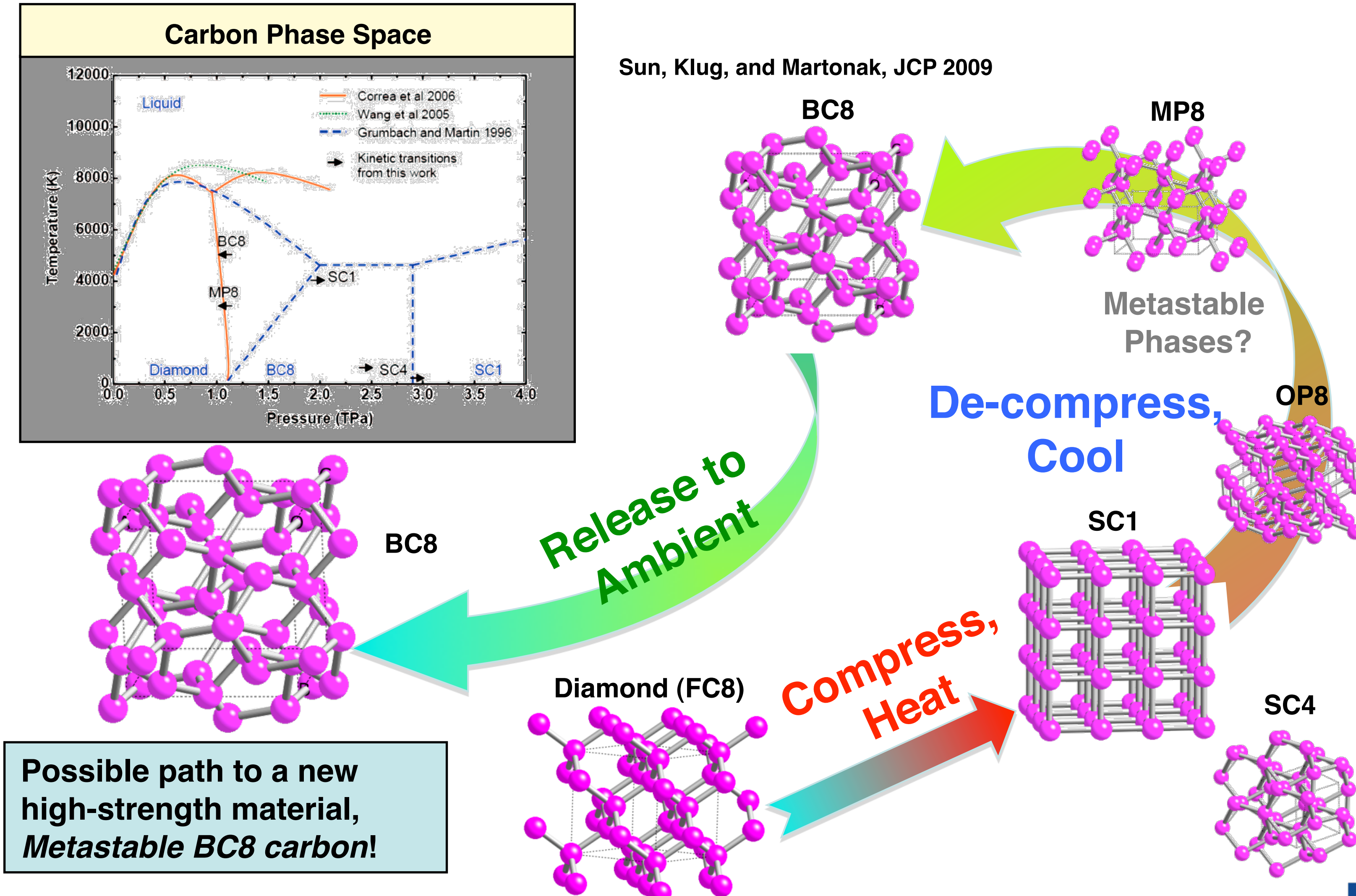
EXAFS offers a temperature probe in multi-shocked iron. BCC phase has not been observed.



This is the first data set at Earth core conditions.

Wide variety of upcoming experiments

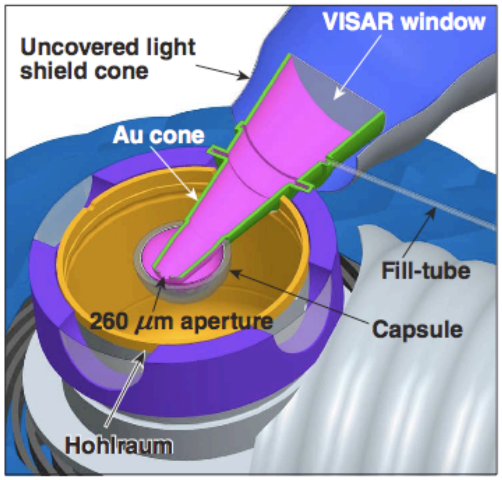
Recent metadynamics survey of carbon proposed a dynamic pathway among multiple phases

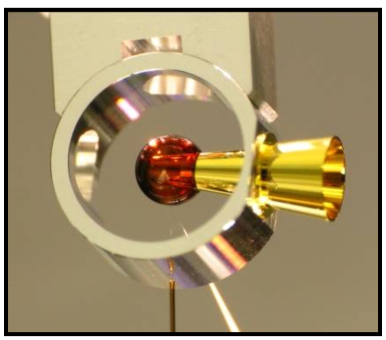


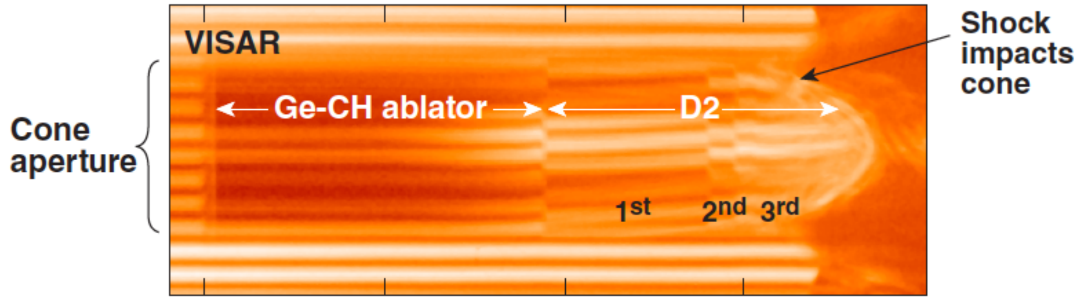
Possible path to a new high-strength material, ***Metastable BC8 carbon***!

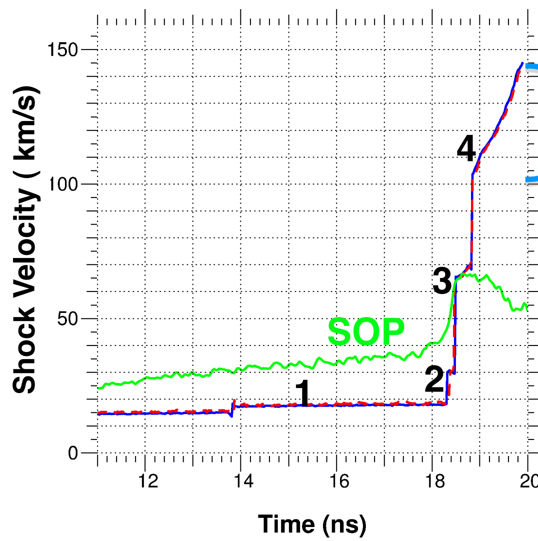
Hydrogen

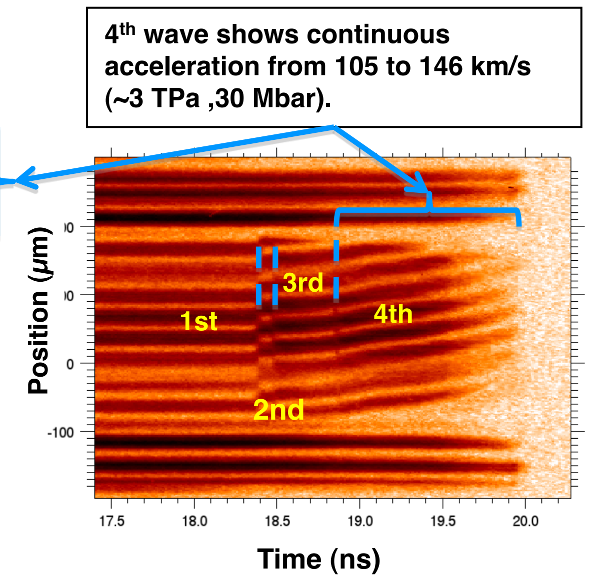
We have tracked shocks in D_2 at 30 Mbar pressures on NIF. We plan to measure the stress-strain of those states.







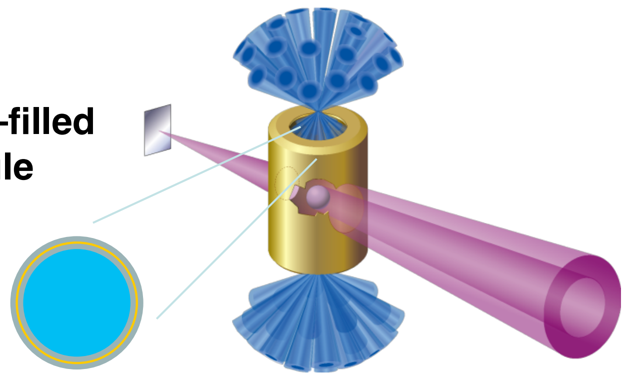


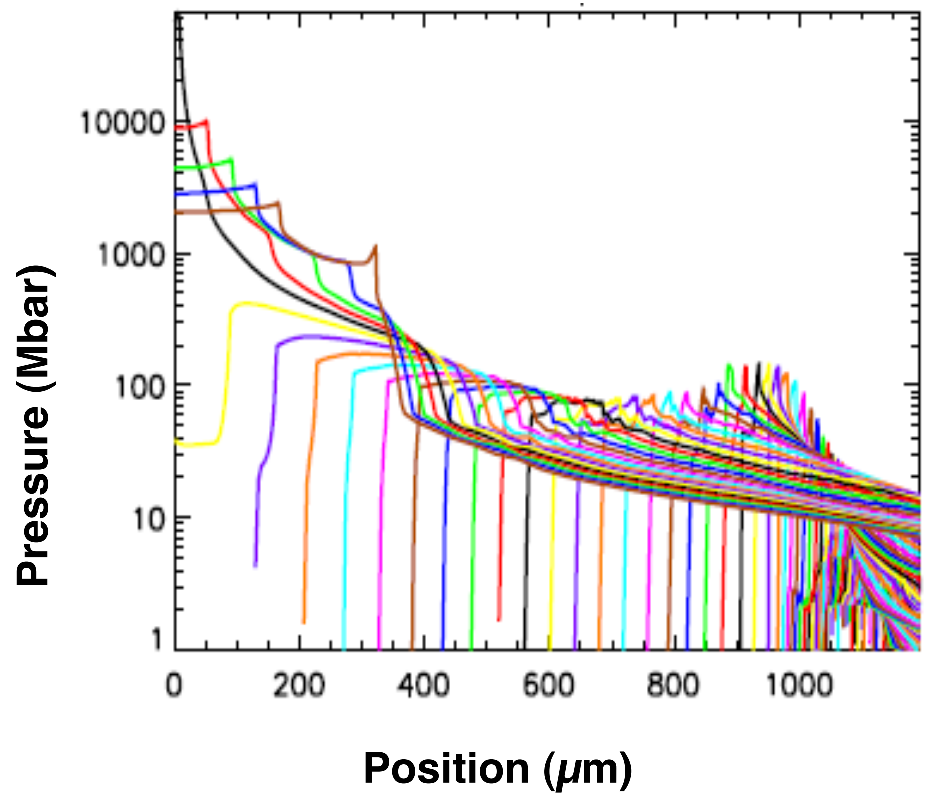


NIC Shock –Timing Work-in-progress with T.R. Boehly, H.F. Robey, J.H Eggert, D.G. Hicks, R.F. Smith, G.W. Collins and many others

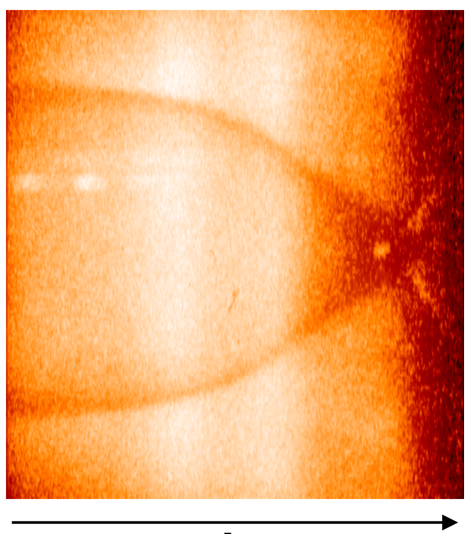
On Omega, we have developed a platform that will allow Gbar pressures on NIF

Liquid D_2–filled NIF capsule





Streaked Radiograph



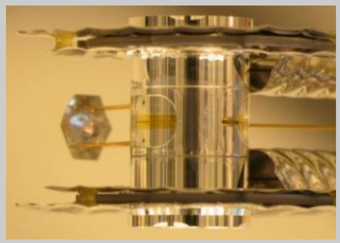
time

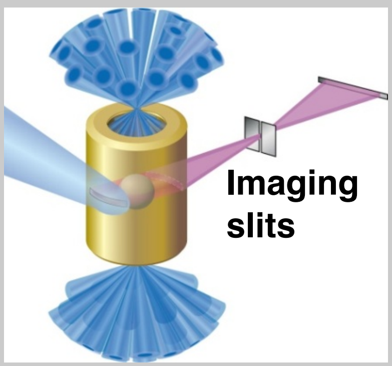
Radiographic probe of density as well as shock velocity

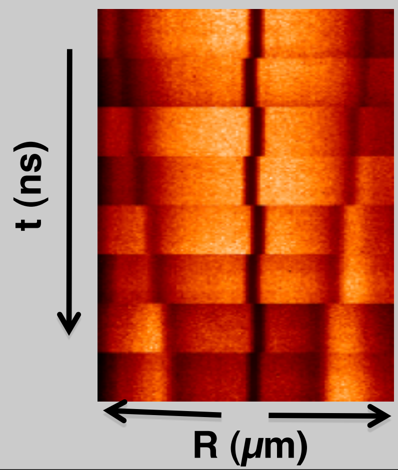
NIF convergent ablator experiments lead to Gbar EOS platform on NIF

Backlit D-³He-filled capsule or THD Cryo-layered capsule

Gated 9 keV X-ray radiography

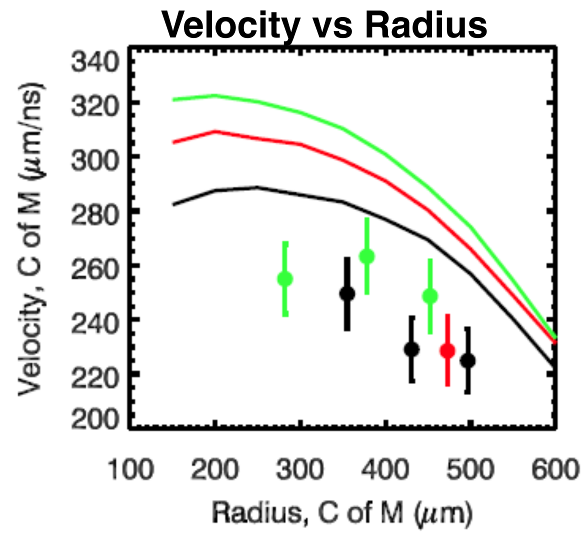


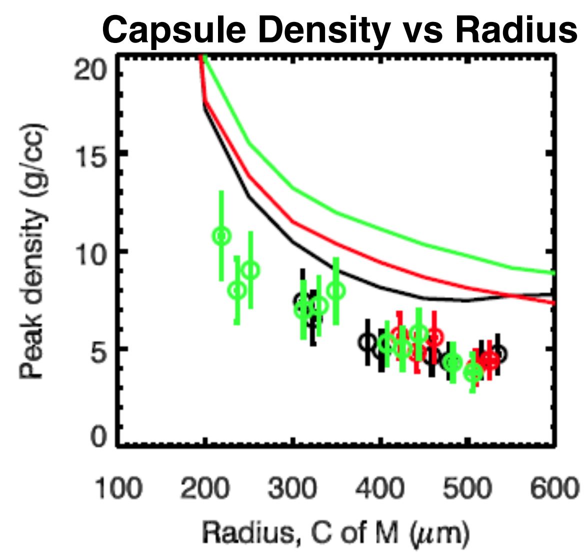




Technique measures radius, velocity, ρR, and mass of ablator

Damien Hicks, Converging Ablator






About 30% of NIF's capacity was needed to reach 50 Mbar on ramped diamond, and about 8% of capacity to reach 9 Mbar in Ta.
We continue to develop experimental techniques to probe these material states:
Stress-Strain
X-Ray Diffraction
EXAFS
Shock Hugoniot
Converging Geometry
Many Others